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Lead-210 as a tracer for acidic deposition in areas of complex topography

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Abstract

This thesis reports an investigation into methods of determining the long term deposition field for atmospheric aerosols in areas of complex topography using the soil inventories of atmospherically derived radionuclides. Measurements of the radionuclides ^{210}Pb, ^{137}Cs, ^{134}Cs and ^{7}Be in vegetation and soil have been made at five mountain locations in northern Britain. A description of the field sampling procedure, sample processing and γ-ray analysis is given. Loss-on-ignition experiments to determine the organic fraction of sampled soils were also conducted on selected samples.

The presence of frequent low level cloud shrouding mountain tops in the uplands of northern and western Britain leads to enhanced precipitation and also deposition of the major acidic ions, eg SO_4^{2-} and NO_3^-, due to the washout of the low level cloud by falling rain ('seeder-feeder scavenging') and also the direct deposition of cloud droplets ('occult deposition'). It is important to quantify the inputs of acidic deposition in these acid sensitive regions and direct measurements present many logistical problems. The radionuclide ^{210}Pb is attached to the same sub-micron aerosol size range in the atmosphere as the major acidic ions and following deposition attaches very strongly to organic matter in soils. In this study the soil inventory of atmospherically derived ^{210}Pb is used as a tracer of the deposition of acidic ions. As the half-life of ^{210}Pb is 22.3 years the soil inventory of ^{210}Pb(atmos), at a site undisturbed for $\sim$100 years, represents deposition integrated over several decades.

Initial sampling took place at the Merrick, an isolated mountain close to the coast in southwest Scotland, to test and develop the technique. Measurements showed the ^{210}Pb(atmos) inventory to increase with altitude at a greater rate than precipitation.

Sampling at Great Dun Fell in Cumbria enabled the measured radionuclide inventories to be compared with detailed measurements of the variation of the wet deposition pattern with altitude, and also with a model of sulphate deposition.

Close correspondence was found between the ^{210}Pb deposition profile and the deposition pattern, with altitude, for the major acidic ions.

The three mountains Ben Cruachan, Beinn Dorain and Ben Lawers lie along an 80 km transect running roughly west to east from the western coast into the central Highlands. Sampling at these three mountains has yielded information on the change in the wet deposition field with distance inland. The measurements suggest that deposition of ^{210}Pb decreases, with distance from the western coast, at a greater rate than does precipitation.

The soil inventory of ^{210}Pb(atmos) increases with altitude at a greater rate than rainfall at 4 of the 5 mountains. The results show that, around summits, on average, the concentration of ^{210}Pb in low level cloud is a factor of ~ 2 greater than in the frontal ('seeder') rain falling from higher altitude. This finding is in good agreement with detailed wet deposition measurements.

The ^{210}Pb measurements are important in themselves as they help define the global and UK ^{210}Pb budget. From measurements made at 65 individual sites the mean ^{210}Pb(atmos) inventory is $0.44 \pm 0.03\,\mathrm{Bq\,cm^{-2}}$, with the mean of the average annual rainfall being $2{,}060 \pm 70\,\mathrm{mm\,yr^{-1}}$. These figures correspond to a mean concentration of ^{210}Pb in rainfall of $66\,\mathrm{mBq\,l^{-1}}$.

The ^{137}Cs inventory has been separated into the fraction originating from nuclear weapons testing, ^{137}Cs(bomb), and the fraction due to the Chernobyl accident, ^{137}Cs(Chern). The measured ^{137}Cs(bomb) inventory ranges from a mean of $3{,}300\,\mathrm{Bq\,m^{-2}}$ at Ben Lawers to $5{,}200\,\mathrm{Bq\,m^{-2}}$ at Beinn Dorain. The ^{137}Cs(Chern) inventory is highly variable between locations ranging from a mean of $440\,\mathrm{Bq\,m^{-2}}$ at Great Dun Fell to $14{,}900\,\mathrm{Bq\,m^{-2}}$ at Ben Lawers reflecting the patchy nature of deposition, due largely to convective storms, during the passage of the radioactive plume. The radionuclide ^{137}Cs is shown to be relatively mobile in the organic soils which characterise the study areas.

The cosmogenic radionuclide ^{7}Be was measured in samples collected from the Merrick on 26th Oct 1988. The mean inventory was $0.039\,\mathrm{Bq\,cm^{-2}}$, being associated with vegetation and the top few cm of the soil profile.

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I would like to dedicate this document to Clare Anderson.

Contents

Chapter 1

Introduction

Recent research into acidic deposition from the atmosphere began with studies in Scandinavia in the 1950's and 1960's into the chemical composition of rain. Declining fish populations in some lakes prompted research into the causes and concern was directed relatively early on at acidic deposition from the atmosphere [41]. Oden concluded that pollution from Britain and central Europe was responsible for acidification of rainfall [118]. Subsequently changes in ecosystems within the UK have been identified. Table 1.1 lists five lochs in the Galloway region of southwest Scotland, an area lacking large scale local pollution sources. These five lochs have shown an increase in acidity over the last few decades. Marked increases in the acidity of lochs in Galloway have been observed over the period 1920–70 [6]. Analysis of diatoms in lake sediment helps to reconstruct the palaeo-environment of the lake. Different species of diatom dominate in different acid conditions and the range of species present provides a history of the acidity of the lake water. Diatom analysis of Loch Enoch has revealed that the pH of the lake was steady at ~ 5.3 between 1600 and 1840 and that by the 1970's the pH had fallen to 4.3 [121].

Recent studies have highlighted the acidification of rainfall due to the long range transport of atmospheric pollutants and most importantly oxides of sulphur and nitrogen, over Europe [70], and North America [64]. At the regional scale (100–1,000 km) the patterns of rainfall-deposited acidity and associated anions

Loch	Change in pH up to mid 1980's	Since
Loch Fleet	6.6–4.5	1961
Loch Skerrow	6.8–5.7	1961
Loch Garroch	6.6–4.9	1961
Loch Harrow	6.3–5.4	1953
Loch Minnoch	6.1–5.3	1953

Table 1.1: Change in pH at five lochs in Galloway, southwest Scotland, between 1953 and the mid 1980's [121]

SO_4^{2-} and NO_3^-, termed 'wet deposition', are quite well defined. Estimates of wet deposition are made by combining the 'precipitation field' with the 'concentration field' for individual chemical species (SO_4^{2-}, NO_3^-, Cl^-, H^+, Na^+, NH_4^+, Ca^{2+} etc). The precipitation chemistry network is low density relative to the high density network of precipitation collectors for rainfall amount [71]. For example, in the UK, the wet deposition estimates are derived from $\sim$4,000 precipitation amount collectors but only 59 precipitation chemistry collectors [86]. A consequence of the small number of precipitation chemistry collectors is that only broad regional concentration patterns can be resolved and some features of the topography of the country are not adequately sampled. One important area of uncertainty is that of the uplands of northern and western Britain. For practical reasons, precipitation chemistry collectors are located at low altitude sites. In estimating wet deposition amounts on hills, the concentrations of the major ions are assumed to be constant with altitude. With this assumption, the uplands of Britain, which experience the largest annual rainfall amounts in the country, receive the largest inputs of wet deposited SO_4^{2-}, NO_3^-, NH_4^+ and H^+ [4][86]. These areas are also biologically very sensitive to the effects of freshwater acidification as a result of the limited base cation buffering capacity of the soils [151].

The enhancement of rainfall amount in these high altitude areas is largely a result of the washout of hill (orographic) cloud by rain falling from above. These orographic clouds frequently shroud the summits of the moderately sized, 500–1,000 m high mountains in the north and west of the UK. This washout of orographic cloud is known as the seeder-feeder scavenging process [9]. Studies have shown that orographic cloud often contains concentrations of the major ions greater than those in rain collected in the same area [45][57][137]. Under favourable conditions, with rainfall falling from higher altitude, cloud droplets may be scavenged very efficiently, resulting in enhanced ion concentrations in the precipitation. In addition to seeder-feeder scavenging, the impaction of cloud droplets against vegetation surfaces may also enhance the deposition of water and of the major ions. Cloud droplets are not collected efficiently by conventional rain gauges and the input due to impaction on to vegetation is termed 'occult deposition' [115]. As a result of these effects of mountain cloud, the wet deposition amounts at high altitude in the UK are larger than the estimates made assuming that rainfall composition remains constant with altitude. Clearly, in these acid-sensitive areas, it is desirable to quantify the effect of mountain cloud on wet deposition levels and to gain a clearer understanding of the mechanisms of deposition. However, the cost and impracticality of working in these remote upland areas means that it is unfeasible to operate a network of precipitation chemistry collectors of sufficient density to monitor wet deposition and occult deposition.

1.1 Aims of study

Recent studies have suggested that radioisotopes in the atmosphere attach to the same size aerosols as the major ions and the inventories of certain radioisotopes in soils may provide valuable information on the scavenging of aerosols from the atmosphere [2][72] [73][112][117]. Graustein & Turekian suggested that the rate and vertical profile of aerosol scavenging may be deduced from inventories of ^{210}Pb and ^{137}Cs in the soil [73]. ^{210}Pb originates from the ground surface as a

daughter product of ^{222}Rn and has a half-life of 22.3 years. Thus, the pattern of ^{210}Pb in soil may provide an assessment of the aerosol scavenging pattern with altitude integrated over about 100 years (ie ~ 5 half-lives). This is in marked contrast to the 'snapshot' taken by atmospheric chemistry experiments which operate over single meteorological events, of typically a few hours duration. It is therefore possible that the measurement of atmospherically derived ^{210}Pb in soils may enable improved estimates of the deposition pattern with altitude in mountain regions to be made.

With this aim in mind soil and vegetation sampling was carried out at five mountain sites in northern Britain, see Figure 3.1. Initially, the Merrick, a mountain with a relatively simple topography, was chosen to test the technique. Later, sampling at Great Dun Fell allowed the ^{210}Pb results to be compared with the acidic deposition observations made at this mountain. The final series of measurements were made on the three mountains, Ben Cruachan, Beinn Dorain and Ben Lawers, which lie along a transect line, which extends ~ 80 km, from the west coast of Scotland, near Oban, eastwards into the central Highlands. Sampling along this transect has produced data on the change in the absolute values of the ^{210}Pb inventory with distance inland and the change in orographic enhancement with distance from the coast. Beinn Dorain and Ben Lawers are located within the Highlands and measurement at these mountains enables the effect of surrounding high ground on the deposition pattern to be investigated. This effect was considered for the rainfall pattern over Snowdonia by Pedgely [123], and for wet deposition of the major ions over Cumbria by Dore et al. [46].

As well as being used as a tool in acidic deposition research, a study of the radioisotope inventories in these areas has intrinsic interest. A large number of values of the ^{210}Pb inventory in soils have been obtained which help to define the atmospheric ^{210}Pb flux to the UK and are useful in developing and modifying models of the global ^{210}Pb and ^{222}Rn budget, see for example [145]. Soil inventories of ^{137}Cs and ^{134}Cs have also been measured, allowing the separation of the ^{137}Cs inventory into the contribution due to weapons testing and that derived

from the Chernobyl reactor failure. Deposition of fallout from Chernobyl was seen to be highly variable between the mountain locations surveyed in this study. Measurements have also been made of the ^{7}Be inventory at the Merrick.

1.2 Emissions of Acidity Precursors

The main anthropogenic precursors of acidic deposition are the gases SO_2, NO and NO_2 (NO and NO_2 are together known as NO_x). Traditionally, SO_2 has been a greater source of acidity than NO_x emissions but the relationship is now changing. Emissions of SO_2 are falling in the UK whereas emissions of NO_x are rising [86]. On a global scale, the contributions of natural and man-made sulphur are roughly comparable. Möller estimates the global natural sulphur emission to be between 50–100 Mtonnes S yr^{-1}, whereas the estimated anthropogenic emission is 60–80 Mtonnes S yr^{-1} [109]. Global NO_x emissions have been estimated as between 25–99 Mtonnes N yr^{-1} with natural sources contributing about a third of this total [104].

The man-made S contribution is produced largely in specific areas, with two thirds originating in Europe and North America. Europe was responsible for producing 44% of the world total of SO_2 emissions in 1979 [150]. Over Europe and North America 'natural' SO_2 and NO_x emissions are insignificant compared with the anthropogenic emissions. In the UK, emissions of SO_2 peaked during the late 1960's and had fallen by 40% by the early 1980's. Since 1983 emissions have remained steady at about 1.9 Mtonnes yr^{-1} of which 70% originates from the burning of coal and oil in power stations [86]. The spatial pattern of emissions of SO_2 has changed in the UK since the 1950's. Emissions have been localised with the building of large power stations, the closure of smaller stations in urban areas and the marked decline in domestic emissions as a consequence of smoke controlled zones. The average height of release has also increased with the use of tall stacks at these large power stations (termed LCP's, Large Combustion Plants). As a result of these changes, the southeast of England, including London, has

declined in relative importance as a source region and the north Midlands and south Yorkshire have increased in relative importance with the construction of several LCP's in the Trent Valley and south Yorkshire. On the European scale a broad band of high emission stetches from this region of the UK, through Germany and into central Europe.

Nitrogen oxides are emitted by power stations and motor vehicles and, as well as being a direct acidity precursor, they also have an important role in the atmospheric photochemical reactions which transform primary to secondary pollutants [75]. Anthropogenic NO_x are mainly produced during the high temperature combustion of fossil fuels, oxidising nitrogen in the fuel and also nitrogen in the air. Total UK emissions of NO_x are around $0.75\,\mathrm{Mtonnes\,N\,yr^{-1}}$ [51]. Motor vehicles are the largest source and continue to grow in importance, although power stations are also a major producer. The spatial distribution is somewhat similar to that for SO_2 emissions though not so localised (as motor vehicles are the major source). NO is emitted from soil [65], and though it is difficult to quantify due to few measurements, if a flux of $10\,\mathrm{ng\,m^{-2}\,s^{-1}}$ is typical of rural Britain then annual emissions of NO from soil may amount to $\sim 10\%$ of those from combustion sources [129]. Another major source of NO_x is biomass burning. As a result of the seasonality in the demand for energy throughout the year, emissions of SO_2 are greatest during the winter and least during the summer months. There is little variation in vehicle usage throughout the year and thus the seasonal variation in NO_x emissions is less than that for SO_2 emissions.

1.2.1 Chemical Reactions and Transport

Oxidation of SO_2 and NO_x takes place as the gases are emitted and dispersed from the source. The oxidants, hydrogen peroxide (H_2O_2), hydroxyl radical (OH) and ozone (O_3) are the most important for SO_2 and NO_x. Hydrocarbons derived mainly from motor vehicles, fuel evaporation and natural gas leakage are important in the production of O_3 in the atmosphere. The rate of removal of sulphur compounds from the atmosphere by rain generally exceeds the rate at which SO_2

undergoes dry deposition. The sulphur content of rain and cloudwater greatly exceeds that possible due to simple dissolution of SO_2 from the surrounding air and oxidation mechanisms play an important role in this enhancement. SO_2 may be oxidised in the gas phase and the liquid phase. NO_2 is rapidly oxidised in the gas phase, at a rate ten times faster than that for SO_2. The various oxidation mechanisms for SO_2 and NO_x are outlined in Figure 1.1. The sulphuric and nitric acids produced in dry oxidation may condense on existing particles or form nuclei around which water condenses to form relatively pure acid droplets.

1.3 Atmospheric Particles

The gases, SO_2 and NO_x are emitted into the atmospheric boundary layer, the layer of the troposphere closest to the ground and subject to turbulent mixing due to heating of the ground and the frictional drag of the ground surface on flowing air. The depth of this layer varies but is typically about 1 km deep [86]. SO_2 and NO_x may dry deposit to the ground surface close to the source or become associated with atmospheric aerosols; incorporation into aerosol usually takes place after oxidation. Turbulent mixing in the atmospheric boundary layer dilutes the pollutant and stimulates chemical reactions through mixing with the oxidant-containing surrounding air.

Atmospheric aerosols vary in size from $\sim 0.001\,\mu$m diameter, no more than small clusters of molecules, up to ~ 1 cm in diameter, for example large hailstones, snowflakes etc. Larger atmospheric particles are extremely rare. Aerosols of $\geq 10\,\mu$m diameter usually only exist in moist air with condensed water vapour. In 'dry' air, in the size range 0.001–10 μm, the number concentration is commonly 1,000 to 10,000 cm^{-3} and at clean, remote sites 100's cm^{-3} [148]. Most atmospheric aerosol particles are smaller than $0.1\,\mu$m diameter in the dry state and it is likely that most of these particles are produced by condensation from the vapour state. These particles then grow in size by diffusive coagulation until the growth rate by this mechanism becomes negligible at $\sim 0.1\,\mu$m diameter. The size

Sulphur

Dry oxidation pathway; the major route involves OH radicals,

$OH + SO_2 \longrightarrow HSO_3$

$HSO_3 + O_2 \longrightarrow HO_2 + SO_3$

Typical rate $\sim 1\%\,hr^{-1}$

Wet oxidation pathway; in the presence of cloud droplets,

$SO_2 \longrightarrow SO_2 \cdot H_2O \longrightarrow HSO_3^- + H^+$

$HSO_3^- + \text{Oxidant} \longrightarrow SO_4^{2-} + H^+$

Nitrogen

NO is converted to NO_2 via a reaction involving a reactive hydrocarbon (RH),

$RH + 2NO + 2O_2 \longrightarrow \text{Carbonyl} + 2NO_2 + H_2O$

During daytime the major reaction is as follows,

$OH + NO_2 \longrightarrow HNO_3$

The rate for this reaction is ~ 10 times that for SO_2

At night a reaction is possible involving the nitrate radical

$NO_2 + O_3 \longrightarrow NO_3 + O_2$

$NO_2 + NO_3 \longrightarrow N_2O_5$

$N_2O_5 + H_2O \longrightarrow 2HNO_3$

Low solubility of NO_x precludes an aqueous oxidation route.

Figure 1.1: Major oxidation pathways for SO_2 and NO_x [86].

range 0.1–1.0 μm is relatively stable [148], and has important properties affecting atmospheric deposition.

Sulphuric and nitric acids, produced by oxidation of SO_2 and NO_x respectively, may become incorporated into the atmospheric aerosol. Sulphuric acid may condense on to existing particles or may self-nucleate to form relatively pure acid droplets. Particulate nitrate forms through the heterogeneous reaction between HNO_3 and basic particles and by gas phase reaction with ammonia (NH_3) [86]. The aerosols over Europe and North America generally contain $(NH_4)_2SO_4$, H_2SO_4, NH_4HSO_4, NH_4NO_3 and are ideal cloud condensation nuclei [56].

Whitby [154] gives the median diameter of the sulphate aerosol as 0.48 ± 0.1 μm with a range of 0.1–1.0 μm and other investigations have found similar size ranges, eg [66]. Nitrate aerosol is much more abundant than nitric acid vapour [86].

The pattern of deposition for pollutants, and the mechanisms involved, are highly dependent upon whether the pollutant is in the gas phase or associated with particulate. In the gas phase, deposition rates are highly dependent upon the chemical properties of the pollutant molecules, while for the particulate the particle size is the most important parameter controlling deposition.

1.4 Atmospheric Deposition Mechanisms

1.4.1 Dry Deposition

Dry deposition is defined as the direct deposition of pollutant gases and particles from the atmosphere to vegetation, buildings and the ground surface. Dry deposition rates are quantified by a parameter called the deposition velocity which, although it has the units of velocity, does not actually correspond to a real velocity. The dry deposition velocity of a pollutant species to a surface is given by:

$$V_d \; = \; F_d \; / \; C(z1) - C(z2)$$

V_d is the dry deposition velocity ($\mathrm{cm\,s^{-1}}$), F_d is the flux of the pollutant

($g\,cm^{-2}\,s^{-1}$) and C(z1) − C(z2) is the concentration difference ($g\,cm^{-3}$) between the reference level in the atmosphere and the absorbing surface [29]. The reciprocal of the deposition velocity gives the total resistance to transfer, r_t, which comprises both atmospheric and surface terms. There are three phases in the dry deposition process: turbulent transport through the free atmosphere to within a few mm of the surface; transport through the viscous sub-layer of air that surrounds all surfaces; and then, finally, capture at the surface. Turbulent diffusion brings gases and particles from the free atmosphere close to the surface so that rates of transport are similar for gases and particles and depend primarily on windspeed and the aerodynamic roughness of the ground [86]. The viscous sub-layer is characterised by airflow which is essentially horizontal to the surface, wind velocity is zero and turbulent diffusion is no longer active. Gases cross this layer by Brownian diffusion and subsequent uptake at the surface is determined by the chemical properties of the gas molecules. For example, uptake of SO_2 by plants occurs via stomata and therefore shows a maximum during the day and a minimum by night [56].

For SO_2 the largest values of dry deposition to the UK occur in the industrial Midlands of England with annual inputs of about $3\,g\,S\,m^{-2}$. The overall pattern of dry deposited SO_2 closely follows that of ambient SO_2 concentrations. Measurements of NO_2 deposition show dry deposition velocities in the range 1–2 $mm\,s^{-1}$ and dry deposition is only a major removal mechanism for NO_x in southern England. Nitric acid vapour (HNO_3) is very reactive and is readily deposited on all natural surfaces. However, as air concentrations are small, inputs by this mechanism at most sites contribute only a small amount of the total N deposited [86].

In considering the dry deposition of particles, size is the most important governing factor. Small particles, $< 0.1\,\mu m$, may pass through the viscous sub-layer by Brownian diffusion, as is the case for gases. Large particles, $> 1.0\,\mu m$, posess sufficient weight to sediment out of the atmosphere and settle on surfaces. These large particles also have sufficient inertia to impact directly against obstacles instead of following the streamlines of the flow of air around the obstacle. This mechanism

of deposition is termed impaction. In practise, the number of particles in this size range are few. The intermediate size range 0.1–1.0 μm is particularly important since it is in this range that much of the SO_4^{2-} and NO_3^- is found. For particles of this size, the processes of Brownian diffusion and gravitational settling are very slow and in the absence of rain these particles are not effectively removed from the atmosphere [56]. Figure 1.2 illustrates the variation of deposition velocity with particle size and it can be seen that there is a minimum in deposition velocity in the size range 0.1–1.0 μm.

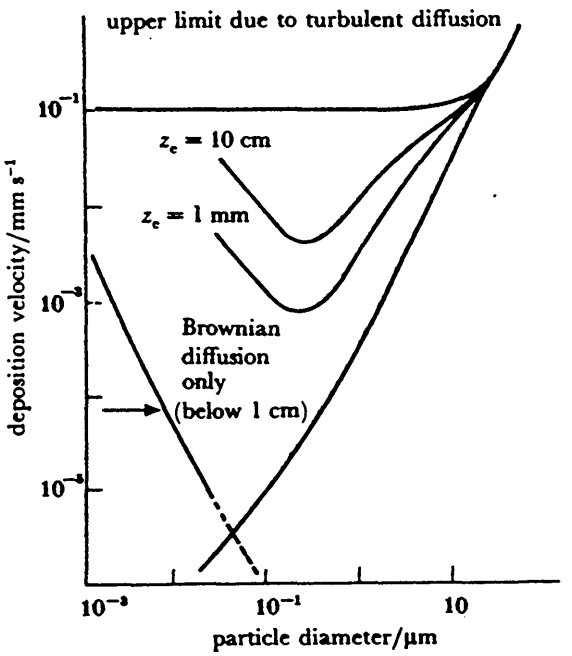

Figure 1.2: The relationship between particle deposition velocity over natural surfaces and particle size for vegetation with two different surface roughness characteristics (z_0). Taken from Fowler et al. [60]

The three stages in the dry deposition process are usually investigated within the framework of the resistance analogy. Within this framework the total resistance to the transfer of gases and particles to the ground, r_t, is separated into atmospheric and canopy terms. Total resistance is given by:

$$r_t = r_{am} + r_{bg} + r_c$$

The term r_{am} represents the aerodynamic resistance for momentum transfer, r_{bg} represents the viscous sub-layer resistance and r_c represents the surface or canopy resistance [60]. A detailed description of the resistance analogy as applied to the transfer of gases between the atmosphere and vegetation is given by Thom [142].

1.4.2 Wet Deposition

The term wet deposition covers a range of processes whereby atmospheric pollutants become associated with hydrometeors and are subsequently transferred to the ground in precipitation. Although studies of precipitation chemistry were made during the nineteenth century, eg [139], it is only in recent decades that systematic observations of rain chemistry have been made in the UK. Wet deposition processes are divided into two groups: processes which act within clouds whereby cloud droplets and growing raindrops scavenge pollutants (rainout) and scavenging of pollutants below the cloud base by falling raindrops (washout). The major process whereby gaseous and particulate pollutants are incorporated into precipitation are, phoretic mechanisms, Brownian diffusion, impaction, solution and oxidation of pollutants in gas form and nucleation scavenging. Phoretic mechanisms and Brownian diffusion are relatively insignificant with their combined contribution to wet deposited sulphur and nitrogen being about 5% for both ions [56].

Impaction of particles against cloud droplets and raindrops occurs when they are too large, ie possess too much inertia, to follow the streamlines of airflow around these hydrometeors. This mechanism is only efficient for particles larger than $\sim 5\,\mu$m [56], and little or no collection is expected below $\sim 1\,\mu$m [148]. Even though this capture mechanism is only efficient for particles $> 5\,\mu$m Fowler [56] estimates, from the work of Garland [67], that this mechanism may be responsible for about 10% of the sulphur and nitrogen in rainfall.

Most sulphur and nitrogen enters precipitation via the nucleation scavenging pathway. The atmospheric aerosols with which SO_4^{2-} and NO_3^- are mostly as-

sociated, typically 0.1–1.0 μm at the humidities and temperatures prevalent over northwest Europe, are hygroscopic and often present as droplets [66]. In subsaturated air, when the humidity changes, these hygroscopic particles rapidly gain or lose water to adjust their size and chemical composition and hence maintain a state of equilibrium [86]. These deliquescent aerosols act as ideal cloud condensation nuclei. Cloud begins to form when the relative humidity of a parcel of air reaches 100%, this is usually the result of moist air rising and cooling by adiabatic expansion. A certain number of these aerosols become 'activated' and continue to grow until supersaturation ceases. Unactivated aerosols may then be slowly incorporated into cloud drops by the collection processes mentioned above. Measurements of particulate sulphate and nitrate at Great Dun Fell, made while cap cloud was present, have shown that the entire SO_4^{2-} and NO_3^- inventory, measured as small particles below the cloud base, may be found in cloud droplets further up the hill [56]. Fowler [56] from the work of Hegg [78] attributes around 65% of sulphur and between 60 and 70% of nitrogen in wet deposition to this mechanism.

Around 20% of sulphur in precipitation is attributed to the solution and oxidation of SO_2 [56]. SO_2 is soluble and exchange takes place between rain and cloud droplets and the atmosphere. Irwin et al. consider that only about 1% of SO_2 is removed by this process, due to the relationship between the solubility of SO_2 and acidity of the atmospheric water. It has been demonstrated that the uptake and oxidation of SO_2 in cloud water by H_2O_2 provides an efficient oxidation pathway for SO_2, eg [37]. Model calculations and measurements have shown that rainout (ie in-cloud) mechanisms such as nucleation scavenging and the oxidation of S IV species are the largest contributors to sulphate in rainfall [7]. The solution and oxidation processes affecting gaseous nitrogen species are less well understood than for SO_2. Fowler [56] estimates that between 15 and 25% of nitrogen in rainfall enters via this pathway. The low solubility of NO and NO_2, in comparison to SO_2, means that a similar oxidation pathway for NO_x within cloud droplets is unlikely. The evidence suggests that nucleation scavenging is the most important mechanism for the incorporation of SO_4^{2-} and NO_3^- into cloud droplets and raindrops,

eg [56][42].

1.4.3 Occult Deposition

Occult deposition is the term used to describe the capture of ground level cloud droplets by vegetation. Early work showed that the occult deposition of cloud droplets may be an important pathway for the deposition of atmospheric pollutants, although before 1980 the focus of attention was towards the regions of the world subject to advected sea fogs and the role of trees as interceptors of this fog, eg [82][92]. In the 1980's, forest dieback in central Europe led to interest in the role of acidic mists, eg [130].

As mentioned earlier the sulphate- and nitrate-bearing sub-micron aerosols (0.1–1.0 μm diameter) do not impact efficiently against obstacles. However these aerosols act as ideal cloud condensation nuclei when boundary layer air is lifted up the slopes of a mountain. For example, at Great Dun Fell these aerosols are associated with cloud droplets of typically 10 μm diameter by the time they reach the summit at an altitude of 847 m asl [31]. and these wind driven cloud droplets are large enough to impact efficiently onto vegetation.

The direct deposition of cloud droplets is unlikely to contribute a major fraction of the water deposited to a hillside [63]. However, cloud commonly contains high concentrations of the major ions compared to rainfall. At Great Dun Fell the ratios of the concentrations of major ions in cloud to the concentrations in rainfall vary from 2 to 8 with mean values closer to the lower value, see Table 1.2

At altitudes of 400 m asl and above in western Britain low cloud is present between 500 and 2,000 hours per year [60]. Dollard et al. state that, at Great Dun Fell, the frequency and duration of low cloud depends critically on height [45]. At an altitude of 560 m, a few kilometres from the summit, cloud is present for a part of 45 days per year while at the summit, 847 m asl, cloud is present for part of 250 days per year. Thus, it is likely that the occult deposition of acidic compounds will be of most significance in the upland regions of the UK where low cloud is relatively frequent and that the amount of occult deposition will increase

Ratio of concentrations cloud / rain				
H^+	NH_4^+	Cl^-	NO_3^-	SO_4^{2-}
3.9	2.4	2.6	2.8	2.0

Table 1.2: Mean of 11 precipitation and cloud events at the summit, spring 1985 [57]

with altitude at these sites.

Dollard et al. measured fluxes of liquid water to the surface at Great Dun Fell and estimated that, at 760 m, occult deposition of sulphur would amount to 20% of that deposited in rainfall. The vegetation was short grass with exposed peat, surface rocks and moorland plants. Harvey measured the flux of droplets to Big Moor, Derbyshire, 330 m asl, and estimated that occult deposition of sulphur amounted to only 8% of sulphur deposited in rainfall [77]. Harvey attributes the difference between his results and those of Dollard et al. to the difference in height between the two measurement sites and cites the work of Lovett [105] who estimated the occult deposition of sulphur and nitrogen at 1,200 m in New Hampshire, USA to be 80% of the wet deposited values. However, the measurements of Lovett were made at a forest site and the efficiency of capture of cloud droplets by forest is much higher than that by short vegetation. Rates of deposition expressed as a deposition velocities vary from between 10 and 40 mm s^{-1} over moorland to > 100 mm s^{-1} over aerodynamically rougher surfaces, for example forests. Gallagher et al. measured the deposition velocities of cloud droplets as a function of their size for the range 2.5 to 15.5 μm (diameter) and found that for the bulk of cloud water the deposition rates to moorland are close to those for momentum transfer.

Irwin et al. describe how estimates of the occult deposition inputs to the UK are derived, taking into account parameters such as, height of cloud base, mean

windspeed and aerodynamic surface roughness for 20 by 20 km grid squares. This approach suggests that the inputs of sulphur and nitrogen by occult deposition contribute between 1 and 30% to the total inputs with the largest inputs being made on the coastal uplands of northern and western Britain [60]. Inputs to low ground are trivial [86].

1.4.4 Deposition at a site

Table 1.3 gives a breakdown of the total deposition of sulphur and nitrogen at one of the 59 precipitation chemistry monitoring sites. The values were calculated by Irwin et al. using the method of Fowler et al., [58]. Eskdalemuir is a moorland site in the Scottish borders, remote from local sources of pollution and similar in many respects to the sites sampled as part of this study. Wet deposition accounts for 80% of the sulphur deposited at this site but only 44% of nitrogen input. Dry deposition of NO_2, HNO_3 and NH_3 accounts for 56% of nitrogen deposited to the site. Occult deposition is seen to be insignificant for both nitrogen and sulphur.

Deposition Mechanism	Sulphur $g\,m^{-2}\,yr^{-1}$	Nitrogen $g\,m^{-2}\,yr^{-1}$
Wet	0.9	0.7
Occult	0.03	0.04
Dry	0.2	0.9
Total	1.1	1.6

Table 1.3: Atmospheric inputs of sulphur and nitrogen to moorland. Eskdalemuir 259 m asl, [58][86]

1.5 Orographic Rainfall

This section will discuss the rainfall pattern in the mountainous areas of western Britain. By superimposing the average annual rainfall map of Britain on to a relief map the very close correspondence between the high rainfall areas and the high altitude regions of Britain is clear. The mean annual rainfall onto Britain and Ireland is shown in Figure 1.3. Pedgley presents two reasons for the higher rainfall and cloud over mountains [123]. Firstly, mountains act as a barrier to moist airsteams which rise, producing cloud by cooling and expansion, and secondly, on sunny days mountains act as high level heat sources which can result in convective cloud formation. Douglas & Glasspoole state that rainfall was classified by Dr. R.H.Mill into three types, orographic, cyclonic (frontal) and convectional, though it was always recognised that the processes were not independent and could act simultaneously [48]. Cyclonic and convectional rain are comparatively irregular in their incidence and their effect becomes smoothed when long periods are considered. On the other hand the orographic effect is overwhelmingly preponderant and the distribution shown on the maps of annual rainfall does not vary greatly from year to year [48].

In the case of the moderately sized hills of western Britain, the enhanced rainfall amounts have been attributed to the effect of the orography on frontal systems coming in from the Atlantic from mainly westerly and southwesterly directions [48][123][135][136]. To give an example of the orographic effect, over the Glamorgan hills the average annual rainfall is roughly 2,500 mm yr^{-1} and over the South Wales coast 1,000 mm yr^{-1}. Seventyfive percent of the rainfall falling on the hills and 60% at the coast is associated with the passage of frontal systems from the southwest quadrant. This corresponds to an average enhancement of a factor of 3 in the rainfall between the coast and the hills during an orographic rainfall event [79].

Douglas & Glasspoole linked the increased rainfall amounts to the effect of orography on the warm sector of frontal systems. The importance of the warm

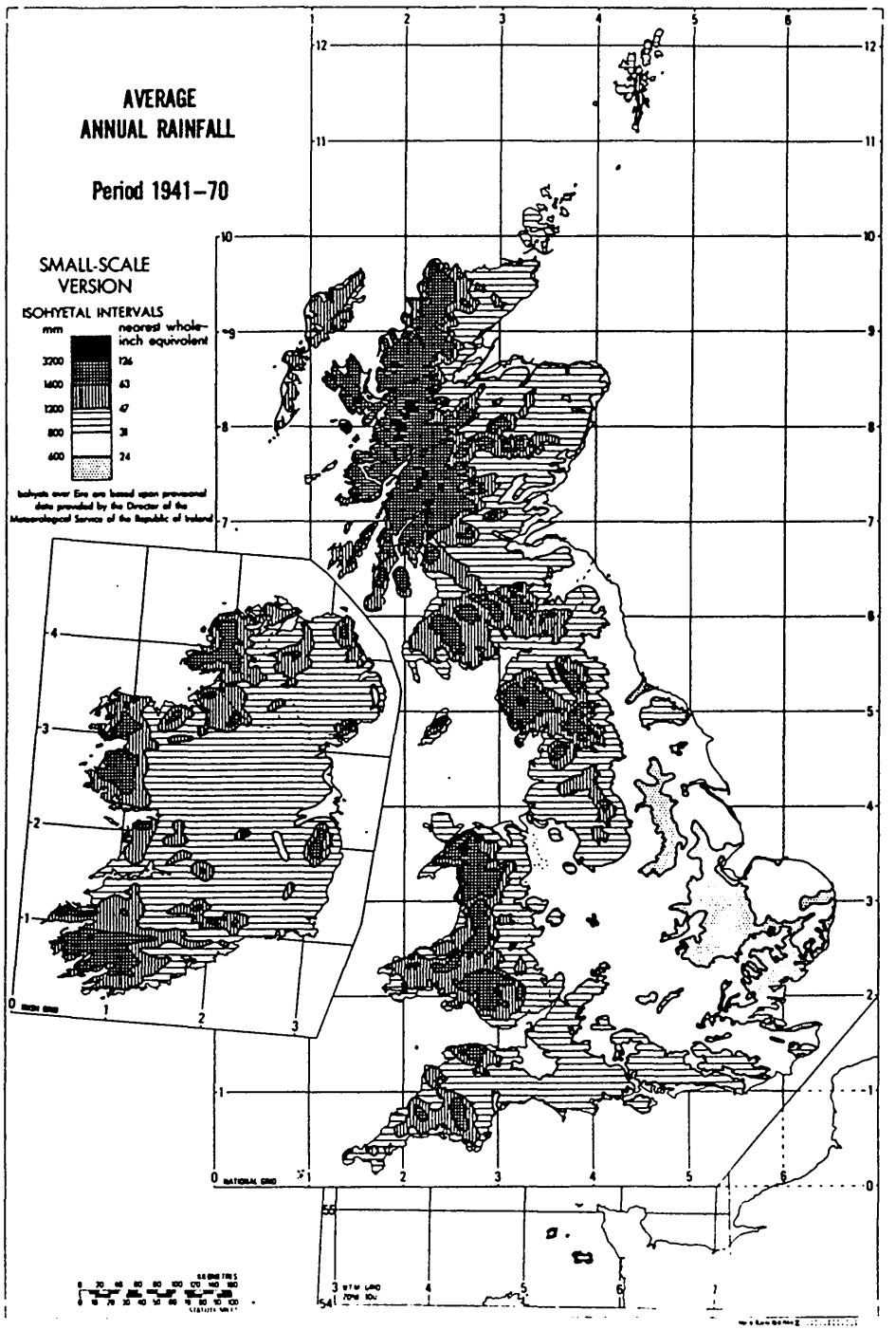

Figure 1.3: Mean Annual Rainfall during the period 1941–70 [108]

sector in producing this extra rainfall has been supported subsequently, for example by the combined autographic raingauge and scanning radar studies carried out by Browning et al. and Hill et al. over the hills of South Wales [17][79]. Browning et al. present a case study of the passage of a frontal system and shows that the rainfall falling from the warm sector is enhanced by a factor of 6 between the coast and the hills and rainfall at the warm and cold fronts was increased by a factor of between 3 and 4.

Another characteristic of orographic rainfall events described by Douglas & Glasspoole, and also emphasised in recent work, is that orography tends to intensify existing precipitation rather than create new areas of precipitation. Although Browning et al. [17] observed the creation of new mesoscale precipitation areas during the passage of the warm sector over the hills, Hill et al. [79] state that the condition of the airmass was more than usually unstable (there was extensive triggering of potential instability). The presence of a deep layer of moist air is important in producing heavy falls of orographic rain [9][48][123].

1.5.1 Seeder–feeder Scavenging

A conceptual model to describe the process whereby rainfall over mountains is enhanced was first put forward by Bergeron [9]. With a very dense network of precipitation gauges on the Uppsala plain operated between 1953–56 and 1958–64 he demonstrated that unexpectedly small orographic features are reflected in the fine stucture of the rainfall distribution. For example, during the autumn months in 1961–63 he observed 40–50 % more rain falling on a small hill than on a nearby lake even though the height difference was only about 50 m. Convection over the hill was ruled out and an orographic effect was suspected.

Bergeron's conceptual model of orographic enhancement incorporated a 'cap' cloud shrouding the hill, formed by the forced uplift of a low level moist airflow. This low level cloud, associated with the hill, in the presence of precipitation from a higher altitude stratiform cloud, is efficiently scavenged of its liquid water. Thus rainfall from above falling through the cap cloud is enhanced by the capture of

cloud droplets. To give an impression of the sizes involved, Bergeron envisaged raindrops of at least 100 μm radius scavenging cloud droplets of typically 10 μm radius. This process he called the seeder–feeder mechanism, the high level stratiform cloud being the seeder cloud and the low level cap cloud being the feeder cloud. The mechanism is illustrated in Figure 1.4.

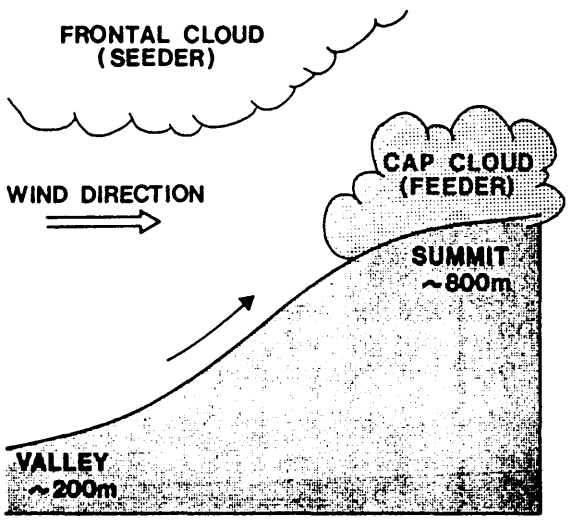

Figure 1.4: The main features of the seeder-feeder mechanism for enhanced rainfall amounts over hills. A medium sized hill is represented

In his model, the seeder rainfall rate, from the stratiform cloud is P_0, P_1 is the rainfall rate falling on the lowlands surrounding the hill and P_2 is the rainfall rate falling on the hill after falling through the cap cloud. The true enhancement due to scavenging of water in the feeder-cloud is given by P_2–P_0, though enhancenment is usually represented by P_2–P_1, as P_1 is easier to measure than P_0. This leads to an overestimate of the enhancement as the rainfall falling from the seeder-cloud outside of the feeder-cloud may be subject to evaporation. Bergeron's conceptual model has been a useful framework, within which a fuller understanding of orographic rainfall enhancement in Britain has been gained.

1.5.2 Studies of Orographic Enhancement

During the 1970's and 80's important work on orographic enhancement mechanisms was carried out over and around the hills of South Wales. Studies incorporating raingauges and scanning radars have revealed that most of the enhancement takes place up to 1.5 km above the hills during the passage of frontal systems and also that orographic rainfall tends to be associated with pre-existing rainfall areas, [17][79].

Theoretical models producing numerical predictions of orographic enhancement due to the seeder-feeder mechanism have been developed. Work by Storebo into the microphysical processes within the feeder cloud revealed that the number density, mean size and composition of condensation nuclei is unimportant in influencing the condensation rate and the resulting rainfall enhancement [141]. Gocho developed a model of rainfall to account for the rainfall pattern around the Suzuka mountains in Japan which experience very heavy orographic rainfall events, usually associated with typhoons advancing from the Pacific ocean [69]. The heaviest rainfall was frequently found to be to the lee-side of the mountains. Only when he incorporated seeding of low level cloud and the horizontal drift of falling precipitation (wind-drift) did his model account for the observed enhancements and the fact that the rainfall maxima occurred to the lee of the mountains. Bader & Roach developed a washout model to account for the rainfall pattern over the South Wales hills [1]. Although their model neglected the effects of wind-drift, this is not significant for the relatively long hills of South Wales, of half-length, L, ~ 20 km as opposed to the 'short' Suzuka mountains of L ~ 4 km.

A model which may be used for 'short' hills was developed by Carruthers & Choularton [24]. Using the basic washout model of Bader & Roach they modified the model to include the effects of wind-drift and stratification. Stratification of the airflow was found to be insignificant in most cases but wind drift was found to move the position of maximum enhancement downwind for both long and short hills. On long hills the maximum enhancement is expected upwind of the hill

crest whereas on short hills it is expected to occur downwind of the summit. Over short hills the maximum liquid water content of the feeder-cloud occurs over the crest of the hill and the greatest enhancement in raindrop size will take place with the raindrops which pass through this region. These drops will reach the ground downwind of the summit as a result of wind-drift. The main factors which influence the degree of enhancement over hills subject to seeder-feeder scavenging are; the initial rainfall rate P_0, windspeed u_0 which influences the condensation rate within the feeder-cloud, hill height h, hill length l and the depth of the feeder-cloud z_c [24]. Other important factors are the moisture content of the low level airflow, ie the relative humidity, and the wet bulb temperature, θ_w, see for example [18]. Two main factors determine the deposition pattern, firstly the path length of the seeder rain drops, related to the hill length and wind velocity, and secondly the rate at which water depleted by the washout process is replaced by condensation [32].

For the moderately sized mountains of the northern and western UK, rising up to $\sim$ 1,000 m asl, there is insufficient time for cloud droplets in the feeder-cloud to coalesce to the minimum raindrop size, $\sim 100\,\mu$m diameter, and then fall to the ground under their own weight [136]. Thus, the cloud droplets are only precipitated in the event of rainfall from higher altitude scavenging the feeder-cloud. Pedgely states that, over the Snowdonian 'dome', air is lifted about 600 m resulting in maximum liquid water contents of about 1 g m^{-3} over the centre of the dome [123]. Large drizzle and small raindrops, ie drops of about 200–1,000 μm diameter, are most effective in scavenging the feeder-cloud. It should be emphasised that the above discussion is only applicable to moderately sized mountain areas which are insufficiently high to disrupt the airflow significantly and which are subject to the passage of moist frontal systems. Good examples include the mountain areas of western and northern Britain and western Norway.

1.6 Acidic Deposition in Mountain Areas

As mentioned earlier, the pattern of wet deposition for the UK is derived by combining the rainfall amounts from $\sim$4,000 precipitation amount collectors with the mean precipitation composition values obtained from the national network of 59 precipitation chemistry samplers. These results indicate that wet deposition of acidity is greatest in the high altitude regions of northern and western Britain. These areas are among those subject to the highest levels of acidic deposition in Europe.

The estimates of wet deposition in the high altitude regions have been derived using precipitation concentration values obtained from low altitude valley sites. However, the assumption has been made that precipitation concentration remains constant with altitude. It has been shown that low level cloud often contains concentrations of the major ions greater than those in rainfall in the same area, eg [57][137]. For the moderately sized mountains, 500–1,200 m of the UK, low level cloud is frequently present; for example cloud is present for a part of 250 days per year at the summit of Great Dun Fell [45]. This cloud may reach the ground surface via occult deposition or by seeder-feeder scavenging. Seeder-feeder scavenging of low level cloud is thus likely to lead to enhanced concentrations of major ions in the falling rain. The above discussion points to orographic cloud as an enhancer of acidic deposition over the mountains of the UK. In the following section, studies of the deposition of chemical species at a mountain in northern Britain are discussed.

1.6.1 Great Dun Fell

For a number of years collaborative experiments including the Institute of Terrestrial Ecology (ITE), the University of Manchester Institute of Science and Technology (UMIST), the University of East Anglia (UEA), Harwell and other research establishments, have taken place in and around Great Dun Fell investigating rainfall, atmospheric chemistry and atmospheric deposition processes. During the au-

tumn of 1984 and the spring of 1985 experiments were carried out to investigate the influence of altitude on rainfall composition and wet deposition. Measurements of rainfall were made at eight different levels (244–847 m) on the western slopes of the hill, between the broad Eden Valley ($\sim$250 m asl) and the summit (847 m asl). Fowler et al. present the results from 20 precipitation events taken when the flow of air was from the west or southwest and orographic cloud was present [57]. On average rainfall at the summit exceeded that in the valley by a factor of 2 and the concentrations of SO_4^{2-}, NO_3^-, Cl^-, NH_4^+ and H^+ in rainfall were larger at the summit than in the valley by factors of between 2.2 and 3.1., see Table 1.4 and Figure 1.5.

Ratio of concentration at summit/ valley for major ions and rain amount					
H^+	NH_4^+	Cl^-	NO_3^-	SO_4^{2-}	Rain
2.9	3.1	2.9	2.3	2.2	2.0

Table 1.4: Ratio of concentrations in feeder/seeder rain and rainfall amount at Great Dun Fell. Values represent the means of 20 precipitation events [57]

Thus, under conditions of west or southwest wind with orographic cloud present it is seen that wet deposition of the major ions at the summit exceeds that in the Eden Valley by a factor of at least 4. The increases in rainfall composition with altitude are attributed to the effects of orographic enhancement which transform particulate pollutants from sub-micron sized aerosols into cloud droplets, typically of mean diameter 5–7 μm [63], as the airmass is lifted up the hillside. These droplets are efficiently scavenged by falling rain from higher altitude, ie the seeder-feeder scavenging, see Figure 1.4. Measurements of cloud water, made at 3 sites (847 m, 808 m and 753 m asl), showed that generally concentrations of the major ions were larger in cloud water than in rainwater by factors of between 1.5 and 8, see Table 1.2. Cloud liquid water content increases with altitude, as the rising air

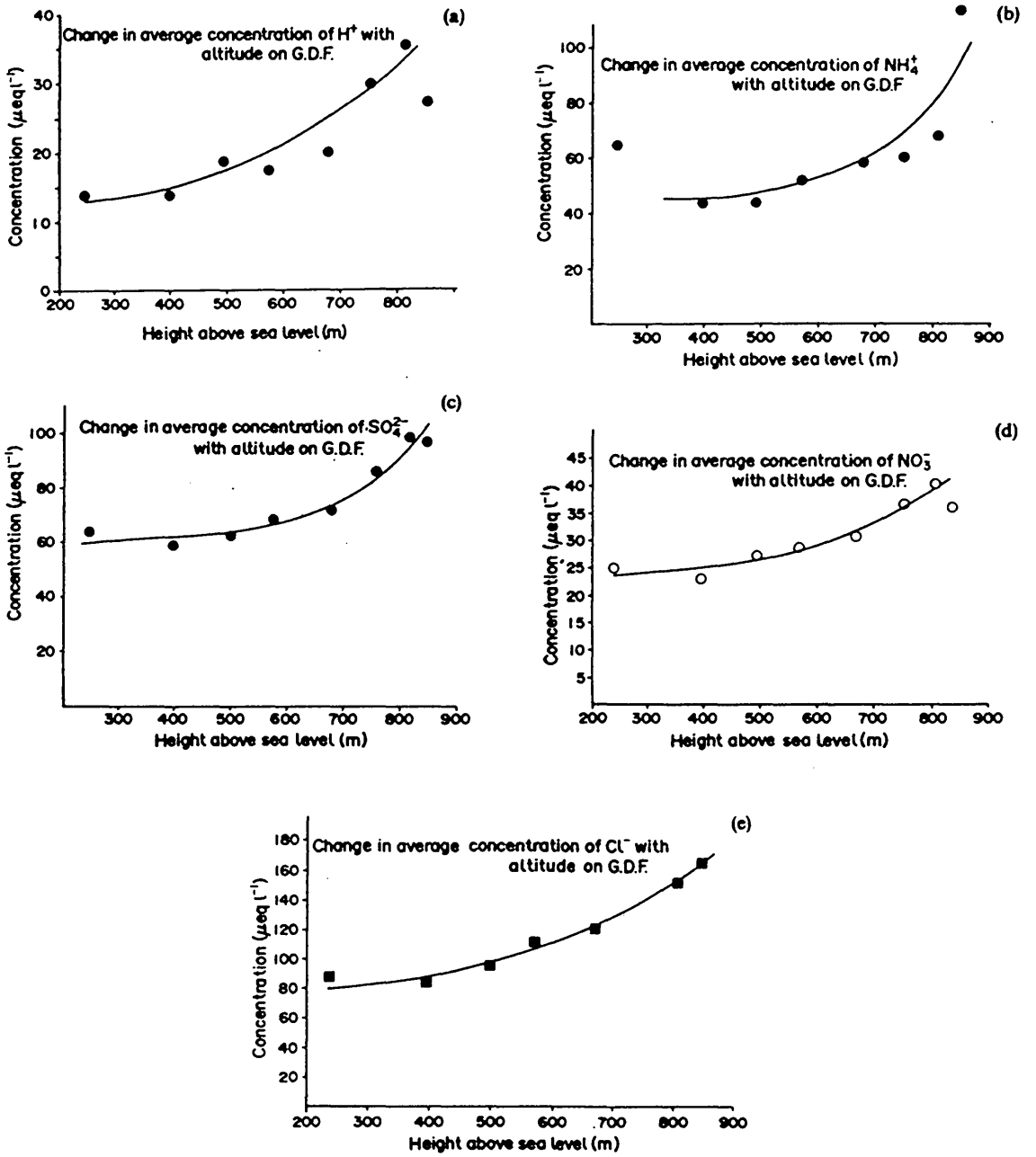

Figure 1.5: Average concentrations, weighted for rainfall amount, of major ions in rain as a function of altitude, 16^{th} April–9^{th} May 1985. (a) H^+, (b) NH_4^+, (c) SO_4^{2-}, (d) NO_3^-, (e) Cl^- [57]

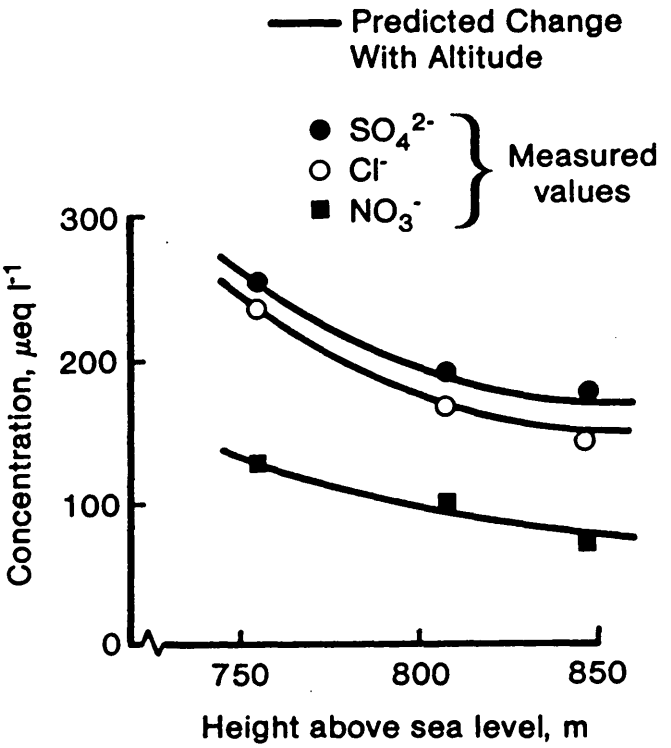

Figure 1.6: The change in concentration SO_4^{2-}, NO_3^- and Cl^- in cloudwater with altitude, compared with that predicted from a calculation of adiabatic water content, Measurements were made on the 17^{th} April 1985 [57]

continues to cool, and this dilutes the solutes present. As a result of the adiabatic growth of cloud droplets, the largest concentrations of major ions are expected at the cloud base, a set of measurements for 17^{th} April 1985 are shown in Figure 1.6.

The seeder-feeder model of orographic rainfall developed by Carruthers & Choularton was modified by Hill et al. to predict deposition rates of sulphate on to hills [80]. Choularton et al. compare the measurements of Fowler et al. with this deposition model [32]. Close qualitative agreement was observed between the model predictions and the measured values for rainfall composition and wet deposition with altitude, although, the quantitative agreement was not so good. At Great Dun Fell, marked changes in both the amount and the composition of rain are observed when orographic cloud is present and a west or southwest wind is blowing. This situation occurs frequently at Great Dun Fell. However, the large changes in rainfall composition are not observed when the wind speeds are light,

ie $< 5\,\mathrm{ms}^{-1}$, and the airflow is blocked upstream. Under these conditions the air at the summit is relatively clean and rainfall will scavenge the more polluted air in the valley. The effect is also not observed when the airflow is from the east [32][57].

Dore et al. report measurements made over an extended region of complex topography [46]. During April 1988 samples of cloud and rainwater were collected at Great Dun Fell and Corney Fell, Cumbria. Corney Fell is situated at the coast, 70 km southwest of Great Dun Fell. Results and model predictions pointed to a gradual 'drying' out of the feeder-cloud as it progressed over successive hills and, in contradiction of the measurements made in 1984 and 1985, the five case studies presented by Dore et al. only found small enhancements of major ion concentrations at Great Dun Fell, both in the field measurements and the model predictions. However, large fluctuations in the major ion concentrations of rainfall occur at Great Dun Fell and the differences between the results of Dore et al. and those of Fowler et al. could be due to the fact that patterns of enhancement are highly variable and strongly dependent on the meteorological conditions [46].

Results do not suggest that the enhanced rainfall compositions observed at Great Dun Fell by Fowler et al. [57] are unique to the site, due to some peculiarity of the topography or meteorology. The effects observed would be expected for most hills on which orographic clouds are common but the rate of change of rainfall composition with altitude depends strongly on hill shape, on atmospheric conditions and on pollution loading of the airmass.

As well as seeder-feeder scavenging leading to enhanced wet deposition on the hill, Dollard et al. estimated that occult deposition of cloud droplets onto moorland at Great Dun Fell would amount to $\sim 20\%$ of the wet deposition amounts recorded in conventional rainfall chemistry gauges alone [45]. From measurements made at 620 m asl on the west facing slopes, Fowler et al. [59] calculated that cloud water deposition to moorland may increase annual wet deposited SO_4^{2-}, NO_3^-, H^+ and NH_4^+ by $\sim 12\%$. It was also calculated that, if the upper slopes of Great Dun Fell were to be afforested, ie an increase in the roughness length from $z_0 \sim 1$ cm to

$z_0 \sim 2\,\mathrm{m}$, then wet deposition amounts would be expected to increase by $\sim 50\%$.

1.7 UK Wet Deposition Estimates

A method to take into account the effects of the seeder–feeder mechanism when producing maps of wet deposition for the UK is described by Dore et al. [47]. Their technique estimates the annual seeder rain along east–west trajectories, taking the rainfall amounts at the east and west coasts and interpolating between them to give the inland seeder rainfall. Any measured rainfall in excess of the calculated seeding values, taken from the 30 year average annual rainfall map [107], represented additional scavenging of cloud water. This cloud water was assumed to contain concentrations of the major ions a factor of 2 greater than those in the seeder rain. Using this technique it was seen that, for land $< 400\,\mathrm{m}$ asl, the orographic enhancement is trivial ($< 5\%$). However, for the very high rainfall areas of Cumbria, Snowdonia, Galloway and the west central Highlands the hilltop grid squares show increases in wet deposition amounts of pollutants by up to 70% in comparison with earlier estimates which assumed a constant concentration of the major ions in rainfall with altitude. These areas of large enhancement include most of the catchments in which freshwater acidification has been identified [151]. It must be said that the approach used here provides only an initial estimate of the orographic effect. Other mechanisms exist for the orographic enhancement of precipitation, for example triggering of instability by the forced uplift of an airmass, and these mechanisms will have different influences on the deposition of pollutants [86].

1.7.1 Total UK Deposition

Estimates of total annual sulphur deposition range from $> 6\,\mathrm{g\,S\,m^{-2}}$ on to the southern Pennines to $< 1\,\mathrm{g\,S\,m^{-2}}$ on to areas of northern Scotland. Estimates of total annual nitrogen deposition are less certain, due largely to uncertainties in the dry deposition behaviour of NH_3, but the national pattern appears to be similar

to that for sulphur and ranges from $\sim 1.2\,\mathrm{g\,N\,m^{-2}}$ for parts of northern England, Wales and Cumbria and $0.4\,\mathrm{g\,N\,m^{-2}}$ for parts of northern Scotland [86]. Recently, it has been suggested that critical loads exist for nitrogen and sulphur deposition. This approach assumes that it is possible to define deposition levels below which ecosystems will not experience adverse effects. Critical loads ranging from 0.3 to $1.0\,\mathrm{g\,S\,m^{-2}}$ for sulphur and $\sim 0.5\,\mathrm{g\,N\,m^{-2}}$ for nitrogen deposition to heathland ecosystems have been suggested by Nilsson & Grennfelt [116]. Deposition is in excess of these suggested loads over much of the UK.

Chapter 2

Radioactivity in Air and Soils

This chapter is concerned with the origin and behaviour of ^{210}Pb, $^{137+134}$Cs and ^{7}Be in the atmosphere, their deposition to the ground surface and subsequent presence in the soil/plant system.

2.1 ^{210}Pb in the Atmosphere

The radioactive isotope ^{210}Pb is a naturally occurring part of the ^{238}U decay chain which begins with ^{238}U (half-life 4.5×10^9 yrs) and ends with the stable isotope ^{206}Pb. Uranium-238 is distributed throughout the earth's crust with typical values of about 33 and 48 Bq kg^{-1} for sedimentary and igneous rocks respectively [30]. The Becquerel (Bq) is the SI unit for the activity of a radioactive substance and is equivalent to one nuclear disintegration per second. The old unit of activity was the Curie (Ci), equal to 3.7×10^{10} Bq, being approximately the activity of 1 g of radium.

The isotope ^{210}Pb is important as an environmental tool, for example as an atmospheric tracer, because a precursor isotope in the chain, ^{222}Rn, is a noble gas isotope which is released from rocks and soils and is free to mix with the atmosphere. Thus ^{222}Rn, along with its daughter products, is redistributed with respect to sources [90]. The ^{238}U series is illustrated in Figure 2.1.

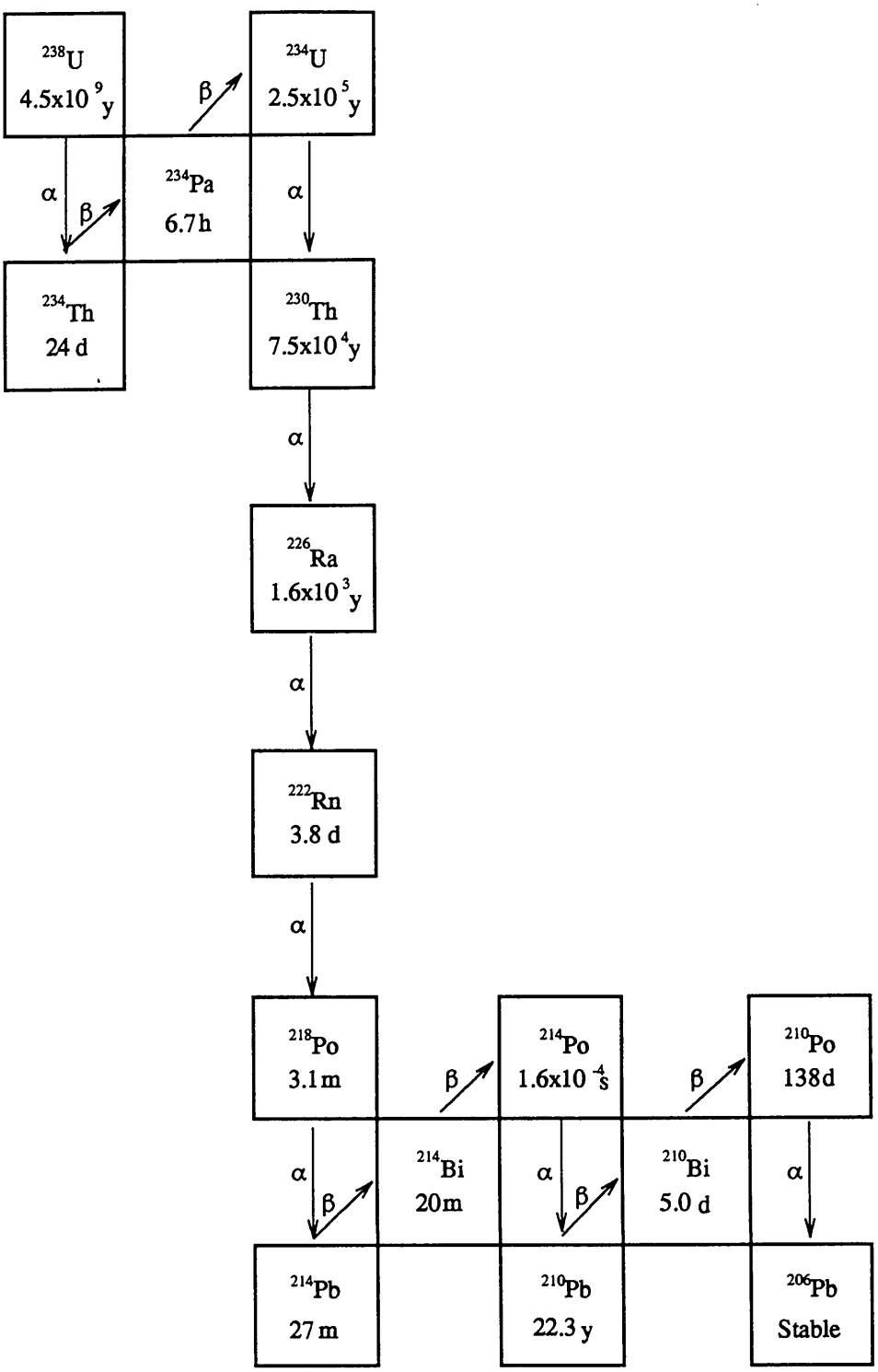

Figure 2.1: The ^{238}U series showing the major decay modes. Each box represents a radioisotope with its half-life

2.1.1 Sources of Atmospheric ^{210}Pb

By far the most important source of ^{210}Pb in the atmosphere is the emanation of ^{222}Rn from the continents. In comparison, the flux of ^{222}Rn from the oceans is small, amounting to only about 2% of that from the land surfaces, even though the oceans cover roughly two thirds of the Earth's surface [132]. All the other natural sources of ^{210}Pb, for example resuspension of soil grains, forest fires and meteoritic and volcanic activity are estimated to provide only about 1.4% of that provided by ^{222}Rn emanation from the land surface. Jaworowski [87] estimated that ^{222}Rn emanation from the land was responsible for 25.5 PBq yr^{-1} production, whilst Pierson et al. [127] estimated 37 PBq yr^{-1} production from all sources (P is a prefix denoting 10^{15}).

Amounts of ^{210}Pb released from anthropogenic sources such as fossil fuel burning, use of phosphate fertilizers and other industrial processes are, on a global scale, minute in comparison with natural sources [88][132]. However, the importance of nuclear weapons testing as a source of ^{210}Pb is uncertain. If a nuclear device contains lead as structural material, ^{210}Pb may be produced on detonation via the reaction $^{208}\mathrm{Pb}\,(\,2\mathrm{n}\,,\gamma\,)\,^{210}\mathrm{Pb}$ [127]. Robbins [132] quotes a value of as much as 52 PBq of ^{210}Pb produced in nuclear weapons tests between 1958 and 1962 from the work of Jaworowski [87]. A concise review of the evidence by Turekian et al. concludes that weapons testing probably did not increase the global ^{210}Pb flux significantly [145].

2.1.2 Production and Diffusion of ^{222}Rn in Soils

The immediate precursor of radon-222 (^{222}Rn) is radium-226 (^{226}Ra). Myrick et al. in 327 samples of surface soil from the USA found a mean activity of 41 Bq kg^{-1} for this isotope [114]. ^{226}Ra is present within the mineral grains and rocks of soils and when this isotope decays, via α-emission, the resulting ^{222}Rn nucleus recoils. If the ^{226}Ra nucleus was close enough to the surface of the mineral grain, the recoiling daughter may come to rest in the space between soil grains. The

fraction of ^{222}Rn which escapes and is free to migrate in the pore space of the soil is dependent upon the surface to volume ratio of the mineral grains and on weathering processes. ^{222}Rn atoms in the pore space may migrate by molecular diffusion and/or flow of the soil gas [34]. Junge [90] assumed the fraction of ^{222}Rn that escaping into the soil air to be about 10% and Baltakmens [2] in New Zealand estimated that between 83 and 93% of radon was retained in soil grains. However, Kurata & Tsunogai [95] estimated that 20–50% escaped in the Japanese soil profiles they studied whilst Barretto et al. measured ^{222}Rn losses of 10–55% from grains [5].

A model presented by Junge [90] describes the production of 'free' radon in soils and its subsequent diffusion through the soil profile and into the atmosphere. In this model it is assumed that the rate of production of ^{222}Rn within the soil, the porosity and the soil density is constant with depth and that the diffusion coefficient has a value of $0.05\,\mathrm{cm^2\,s^{-1}}$. The modelled profile shows the concentration of ^{222}Rn in the soil air to have a minimum value close to the surface and to increase sharply with depth until a constant value is approached at a certain depth, see Figure 2.2.

If radon escape to the atmosphere is by diffusion alone then the fraction of radon escaping from a soil will decreases exponentially with increasing depth in a soil profile [73]. Graustein & Turekian found that, in the upper 20 cm of soils more than 90% of the ^{222}Rn which emanates is lost to the atmosphere. Clements & Wilkening [34] and Nozaki et al. [117] used model calculations, based on diffusion coefficients and the fraction of ^{222}Rn that emanates into the soil air, to calculate ^{222}Rn loss to the atmosphere as a function of depth.

2.1.3 Escape of ^{222}Rn to the Atmosphere

The proportion of ^{222}Rn escaping from a soil depends on a range of factors for example, the moisture content of the soil, whether the ground is frozen or whether there is a snow cover. Junge talks of up to a 70% decrease in ^{222}Rn release during precipitation episodes [90].

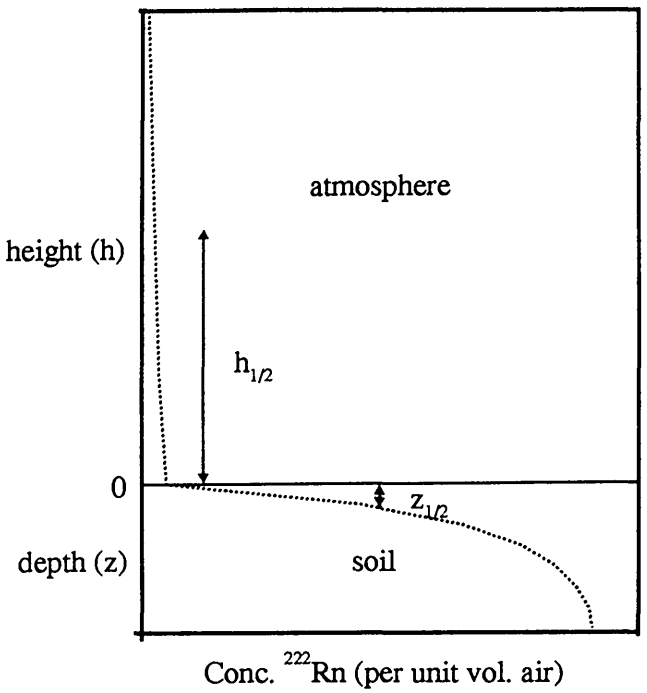

Figure 2.2: A schematic diagram of ^{222}Rn concentration in soil air and in the atmosphere. Typical values deep in the soil profile, c_{s0}, and at the ground surface, c_{a0}, are, $4.8\times10^{-3}\,\mathrm{Bq\,cm^{-3}}$ and $4.8\times10^{-6}\,\mathrm{Bq\,cm^{-3}}$ respectively. Depth where the concentration in soil air falls to $c_{s0}/2$, ie $z_{1/2}$, is $\sim 1\,\mathrm{m}$ and the depth where the concentration in the atmosphere falls to $c_{a0}/2$, ie $h_{1/2}$, is $\sim 100\,\mathrm{m}$ [90]

As well as the molecular diffusion mechanism, other mechanisms have been proposed to explain the ^{222}Rn flux to the atmosphere, for example Clements & Wilkening put forward the mechanism of atmospheric pumping [34]. They observed a 20–60% change in the ^{222}Rn flux associated with an atmospheric pressure change of only 1–2% during the passage of a frontal system. Moreover, soil water contains dissolved ^{222}Rn and transpiration by vegetation may deliver significant amounts of ^{222}Rn to the atmosphere [122].

Commonly quoted values of the ^{222}Rn flux from land are $0.7\,\mathrm{atoms\,cm^{-2}\,s^{-1}}$ [90], and $0.75\,\mathrm{atoms\,cm^{-2}\,s^{-1}}$ [155]. However, Turekian et al. discuss the two most common methods of measuring the ^{222}Rn flux, namely direct measurement with an accumulator and integration of atmospheric ^{222}Rn profiles, and conclude that both methods underestimate the average flux [145]. They used a value of

1.2 atoms cm^{-2} s^{-1} in their global model of ^{222}Rn flux from and ^{210}Pb flux to the Earth's surface.

2.1.4 ^{222}Rn in the Atmosphere

Once ^{222}Rn escapes into the atmosphere it migrates horizontally and vertically by turbulent transport. Migration rates are much more rapid than in the soil profile, for instance, the eddy diffusion coefficient used in the model of ^{222}Rn distribution described by Junge [90] has a value of 5000 cm^2 s^{-1}. As it is a chemically unreactive gas, ^{222}Rn consequently does not attach readily to atmospheric particles. As a result it is almost wholly lost from the atmosphere through radioactive decay. ^{222}Rn (half-life 3.8 days) decays via a number of relatively short-lived daughter isotopes to ^{210}Pb, which has a long half-life (22.3 years) in comparison, see Figure 2.1. Ions of the intermediate daughter isotopes, all with half-lives of less than an hour, possess positive charge and become rapidly associated with atmospheric aerosols.

2.1.5 Size Distribution of ^{210}Pb

The mechanism of attachment of the radon daughters to atmospheric aerosols is analogous to the attachment of SO_4^{2-} ions after oxidation of SO_2 [73], and ^{210}Pb is therefore associated with the same sub-micron particle size range. There is a consensus in the literature pointing out the importance of ^{210}Pb transport on sub-micron sized aerosols. Junge implies an activity median diameter of 0.16 μm for the newly attached, short-lived ^{222}Rn decay products, although with time the particle size distribution will be shifted to larger sizes by coagulation [90]. Turekian et al. state that the mass median diameter of ^{210}Pb containing aerosols is in the range 0.3–0.4 μm and that typically 90% or more of the activity is associated with particles of $< 1\,\mu$m diameter [147]. Measurements of air in rural New Jersey showed that 85% of ^{210}Pb is associated with aerosols of $< 1\,\mu$m diameter [94]. Knuth et al. also present the results of Sanek et al. for Mediterranean air [134],

they found that 70–90% of activity was associated with particles of $< 1.2\,\mu$m diameter. Robbins [132] states that about 90% of ^{210}Pb is attached to aerosols with radii between 0.05 and 0.5 μm and that a maximum abundance is found at $\sim 0.1\,\mu$m.

2.1.6 Concentrations of ^{210}Pb in the Atmosphere

The concentration of ^{210}Pb per unit mass of air over the UK may change with either a positive or negative gradient with altitude. However, the average condition appears to be an increase in concentration with altitude [21][127]. For example, averaged observations for 1958 show an increase from 0.21 mBq kg^{-1} of air at ground level to 0.26 mBq kg^{-1} at 7.6 km and 0.36 mBq kg^{-1} in the stratosphere [127]. The density of the troposphere decreases with altitude, and in terms of concentration per unit volume there is a decrease by a factor of ~ 4 between the ground and the tropopause [90]. Thus, the bulk of the atmospheric inventory of ^{210}Pb is present in the lower half of the troposphere.

The ^{210}Pb concentration per unit mass of air is thought to increase with altitude over the UK due to scavenging and deposition from the lower layers of air masses travelling from east to west over the Atlantic [30]. The source of the stratospheric ^{210}Pb over the UK is thought to be air which ascends from ground level at the equator by convection, then enters the stratosphere and is transported to higher latitudes [21]. The vertical profile of ^{210}Pb over the UK contrasts with that over the continental United States. Moore et al. observed a decrease in ^{210}Pb concentration with altitude to 0.04 mBq kg^{-1} at the tropopause but then an increase to 0.28 mBq kg^{-1} in the stratosphere [111].

Measurements of ^{210}Pb in ground level air reflect the continental effect and concentrations tend to be reduced where the climate becomes oceanic [30]. Greater concentrations are observed in the northern hemisphere than in the southern hemisphere due to the greater land area in the northern hemisphere and the slow rate of mixing across the equator [90]. See Table 2.1.

Reference	Location	Dates	^{210}Pb in surface air ($mBq\,m^{-3}$)
Peirson et al. [127]	Chilton, UK	1961–65	0.23
Graustein & Turekian [73]	9 sites, eastern USA	1985–86	0.5–1.0
Kauranen & Miettinen [91]	3 sites, Finland	1969–70	0.5
Turekian et al. [147]	13 sites, Pacific Ocean		0.033–0.27
Lehmann & Sittkus [96]	Freiburg, Germany	1959	0.37
Bonnyman et al. [15]	Melbourne, Australia	1966–70	0.16

Table 2.1: Mean concentration of ^{210}Pb in surface air at selected locations

2.1.7 ^{210}Pb in Rainfall

Typical values of the ^{210}Pb concentration in rainwater are given in Table 2.2. Blifford et al. observed that, in general, the first part of a rainfall event contained the highest concentration of activity [10]. Fukuda & Tsunogai reported a seasonal variation of the ^{210}Pb concentration in rainfall for Hokkaido, Japan [62]. They also found linear relationships between ^{210}Pb deposition and rainfall amount which showed a concentration 3.5 times greater in winter than in summer due to the northwest monsoon in winter. In general, however, seasonal variations in rainfall concentration have not been observed [145], for example no seasonal variation in concentration was observed in measurements made of rainfall at Milford Haven [127]. Fukuda & Tsunogai concluded from their observations that in-cloud scavenging (ie rainout) was the major removal process rather than below-cloud washout.

2.1.8 Deposition of ^{210}Pb

As ^{210}Pb is associated with the same aerosol size fraction as the ions SO_4^{2-} and NO_3^-, namely the 0.1–1.0 μm fraction, the deposition mechanisms discussed in

Reference	Dates	Location	^{210}Pb in rain (mBq l^{-1})
Burton & Stewart [21]	Sep–Dec 56	Harwell, UK	83
Eakins & Morrison [49]		UK	74
Eakins et al. [50]	Nov 76–Oct 77	Cumbria, UK	70
Clifton [35]*	Nov 88–Nov 90	Plymouth, UK	62±8
Peirson et al. [127]	1962,3	Milford Haven, UK	84
Fukuda & Tsunogai [62]	Feb 70–Mar 72	Hokkaido, Japan	222–300
Knuth et al. [94]	Apr–Jun 77–79	New Jersey, USA	133
Blifford et al. [10]	$\sim$1 year	Samoan Islands	9

Table 2.2: Mean concentration of ^{210}Pb in rain at selected locations. * This value has been calculated by dividing the annual flux of ^{210}Pb in rainfall, given by the author, by the mean annual rainfall shown on the 30 year rainfall map

section 1.4 also apply to the ^{210}Pb carrying aerosols. ^{210}Pb is removed from the atmosphere by wet deposition, dry deposition and, in areas subject to frequent ground-level cloud or fog, occult deposition. The relative importance of these mechanisms for ^{210}Pb deposition has been determined at only a few locations [145]. Knuth et al. measured ^{210}Pb deposition in both wet and dry buckets of a HASL-type collector in New Jersey found 91% of ^{210}Pb in the wet collector [94]. Comparing ^{7}Be and ^{210}Pb deposition in wet-only and bulk samples at Norfolk, Virginia, Todd et al. found that dry deposition accounts for less than 10% of the total ^{7}Be and ^{210}Pb flux to bucket collectors [143]. Graustein & Turekian concluded that dry deposition is only a minor pathway for the delivery of radionuclides to lowlands of the eastern USA [73]. The proportion of dry deposition is expected to increase with altitude due to the greater mean wind speeds at higher elevations. At Graustein & Turekian's forest sites, above 700 m, they estimated that about 58% of the atmospheric ^{210}Pb measured in their soil cores originated in surface air and was removed by dry deposition, seeder-feeder scavenging and occult deposition of cloud droplets.

2.1.9 Residence Time of the ^{210}Pb Aerosol

The mean residence time of tropospheric aerosols has been investigated by measuring the ^{210}Bi / ^{210}Pb and ^{210}Po / ^{210}Pb ratios in air and rain. The degree of disequilibrium between the radionuclides provides a measure of the survival times of the aerosols in the air column relative to removal to the surface [145]. Both ratios overestimate the actual residence times, the polonium couple to a greater degree than the bismuth couple. The polonium couple is very sensitive to injections of stratospheric air and resuspension of soil material, events which produce artificially 'old' values for the tropospheric aerosols. Graustein & Turekian estimated residence times for ^{137}Cs- and ^{210}Pb-bearing aerosols by dividing the tropospheric inventory by the deposition flux to the ground [73].

It appears that the mean residence times of aerosols in the troposphere varies according to location and values for the UK are higher than those for the central

United States. Eakins et al. state that the residence time of ^{210}Pb in the atmosphere over the UK has been reported by various workers, to range between 9 days and a few weeks [50]. Chamberlain [30], from the measurements of Peirson et al. [127], gives a value of 8 days. Moore et al. [111] give a mean residence time of 4 days over the mid-west of the USA and Graustein & Turekian [73] give a value of 4.8 days. The faster removal rate may be due to the fact that relatively more of the ^{210}Pb is in the lower atmosphere over the USA [30]. The increase in residence time between the ground and the tropopause, over the US, has been estimated to be less than a factor of three [73][111]. As a point of interest, Tsunogai & Fukuda found a mean residence time, using the ^{210}Bi couple, of 10–14 days in Hakodate, Japan where the climate is rather oceanic [144].

2.1.10 Measuring the ^{210}Pb Flux

Since the mean residence time of ^{210}Pb in the atmosphere is of the order of a few days magnitude and its radioactive half-life is 22.3 years, very little decays before removal back to the surface. Estimates of the atmospheric ^{210}Pb flux based on measurements of precipitation underestimate the true value since a proportion of the ^{210}Pb is dry deposited. Dry deposition has been estimated to account for about 10% of total deposition over the lowlands of the northeastern United States, eg [94][119]. Continuously exposed, integrating samplers which intercept wet and dry deposition may be expected to provide more representative estimates of the true ^{210}Pb flux [145]. However, due to possible monthly and yearly variations in the ^{210}Pb flux, extrapolation of short measurement periods may lead to significant errors. Fluxes calculated from fewer than 6–12 months' data may be uncertain by a factor of 2 for many places and even several years' data may fix the average flux to no better than 10–50% [145]. In addition, much effort and expense is required to operate atmospheric precipitation collectors, especially in remote areas.

Since the 1960's attention has focused on the use of natural repositories as integrating collectors of ^{210}Pb deposition over many years. The particular subject of this thesis is the measurement of radionuclides in soils and so this field will be

covered in depth. However it should be noted that ^{210}Pb fluxes have also been assayed from; glaciers, ice-caps, lichens, lake deposits and salt marshes and the use of these repositories has been reviewed concisely by Turekian et al. [145]. Dibb from a study of an ice core in Greenland has found evidence that the ^{210}Pb flux may not have been constant over the last 100 years [44].

2.1.11 ^{210}Pb(atmos) in Soils

Fisenne discovered significantly higher levels of ^{210}Pb in the upper 11 cm of undisturbed tobacco-growing soil from Maryland, USA than could be supported from the decay of ^{222}Rn in the soil [55]. Using the model of Junge [90] to account for the ^{210}Pb produced within the soil, she attributed the excess ^{210}Pb to atmospheric deposition and calculated that $\sim 75\%$ of ^{222}Rn remained trapped within the soil grains. The ^{210}Pb produced within the soil is referred to as 'supported' ^{210}Pb and atmospheric ^{210}Pb is known as 'unsupported' ^{210}Pb or ^{210}Pb(atmos). Baltakmens measured the specific activity of ^{210}Pb and ^{226}Ra in 4 New Zealand soils to a depth of 30 cm [2]. Values for total ^{210}Pb varied between 11 and 33 Bq kg^{-1} and those for ^{226}Ra between 15 and 33 Bq kg^{-1}. Baltakmens found that the ^{210}Pb(atmos) remained within the top 15 cm and that the flux estimates were in good agreement with values obtained by direct measurements. A typical soil profile is shown in Fig 2.3.

An association between ^{210}Pb(atmos) and organic matter (OM) in soils has been found, eg [36][97]. Lewis considered that OM in soil was almost 100% efficient at scavenging and retaining ^{210}Pb(atmos) and that soil erosion was the only significant removal process [97]. He also estimated a mean residence time of ^{210}Pb in soils of the order of 2,000 years. Benninger et al. estimated a value of about 5,000 years and concluded that less than 2% of ^{210}Pb in rainfall is transported to groundwater [8]. However, there is evidence that ^{210}Pb is mobilized by the organic-rich waters of ombrotrophic peatlands in the USA where greater amounts of ^{210}Pb are found under hummocks than in hollows [149]. Eakins et al. compared the concentration of ^{210}Pb in rainfall with that entering a lake for a catchment

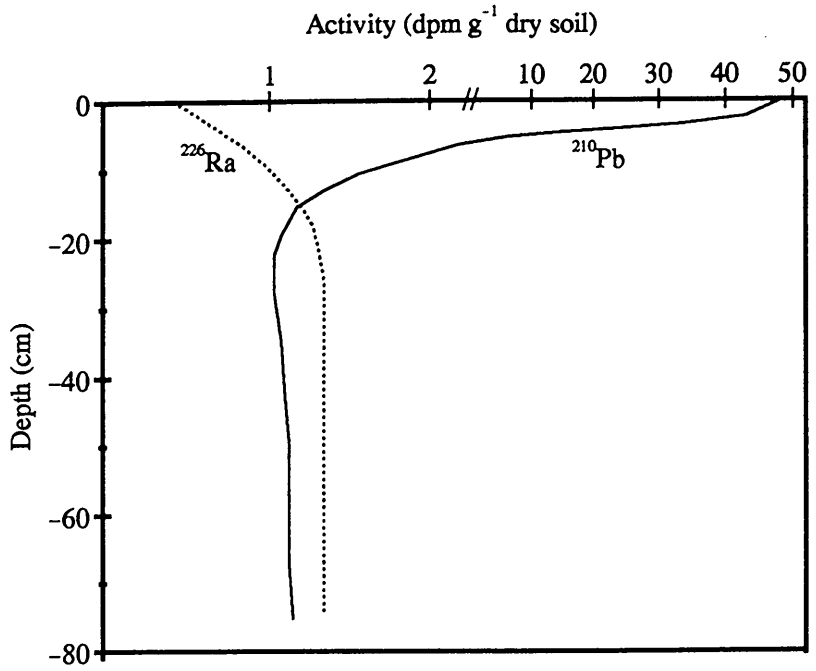

Figure 2.3: Calculated profile of ^{210}Pb and ^{226}Ra for a soil from Cook Forest, Pennsylvania. The excess of ^{210}Pb over ^{226}Ra in the top few cm represents the atmospheric input. The deficiency of ^{210}Pb compared to ^{226}Ra deep in the profile is a result of radon loss. The symbol dpm stands for disintegrations per minute, 1 Bq = 60 dpm. Adapted from Nozaki et al. [117]

in northern England and found that the concentration per litre of water fell by a factor of about 40 between the water falling as rain and its flowing into the lake as stream water [50]. This illustrates the ability of soils to scavenge ^{210}Pb in rainfall.

Linear relationships have been observed between the percentage of OM in soil and the concentration of ^{210}Pb(atmos) per gramme of OM for individual soil profiles [97][117]. In addition Nozaki et al. found this relationship to hold between individual soil profiles on a regional scale for Pennsylvania soils [117]. Lewis considered this relationship to be a result of rapid homogenisation of ^{210}Pb within the OM fraction caused by the burrowing action of animals [97]. Atmospheric deposition rates of ^{210}Pb are obtained most easily from soils in which the OM is concentrated in the surface layers, because where the OM is more evenly distributed, the lower concentrations of ^{210}Pb lead to the measured deposition rates

being more sensitive to errors in the 'supported' ^{210}Pb measurements.

For soil sampling to work, it is necessary to select a site which has been undisturbed for about 100 years; that is, one unlikely to have been affected by erosion, sedimentation, ploughing or animal burrowing. Sites subject to large runoff and accumulation of precipitation should be avoided [112]. In taking a core sample the cross-sectional area and depth interval of each subsample need to be noted so that values of $Bq\,kg^{-1}$ may be converted to $Bq\,cm^{-2}$. Correction for 'supported' ^{210}Pb is made for each depth interval and the total ^{210}Pb(atmos) inventory is calculated by summing the values for each depth interval. The amount of 'supported' ^{210}Pb is usually calculated by measuring the ^{226}Ra activity in the soil, although, as mentioned earlier, this leads to an overestimate of the supported ^{210}Pb fraction because ^{222}Rn diffusion and loss from the soil profile is not accounted for. Several authors have modelled this process and hence estimated the uncertainty arising. Moore & Poet give the activity ratios of ^{210}Pb / ^{226}Ra in surface soils for 9 sites in Colorado, Kansas and Texas [112]. The ratios range from 1.7 to 5.8 for the 0-1 cm depth layers. These values are quite low and could indicate that the soils sampled were sandy/mineral soils with only a small proportion of OM. For the soils analysed the ^{210}Pb(atmos) was confined to the upper 20 cm of soil. The fraction of ^{222}Rn escaping was estimated to be 20% and thus the 'supported' ^{210}Pb fraction was 80% of the measured ^{226}Ra activity.

Nozaki et al. modelled observed ^{210}Pb and ^{226}Ra profiles to correct for ^{222}Rn loss and concluded both that ^{222}Rn loss had a small effect for the soils studied and that use of the ^{226}Ra activity led to an underestimate in the ^{210}Pb(atmos) flux of only about 10% [117]. In contrast Kurata & Tsunogai [95] compared soils which had been exposed to rainfall with soils which had been sheltered underneath houses and concluded that deposition rates estimated without taking account ^{222}Rn loss underestimate the ^{210}Pb(atmos) flux by 10–50% for the soils studied and that correction for ^{222}Rn loss is particularly important for soils with low activity ratios of ^{210}Pb / ^{226}Ra (<10) at the surface. Nevertheless, most authors point to an underestimate of no more than $\sim 10\%$.

The flux of ^{210}Pb(atmos) may be obtained from the soil inventory if the soil has suffered no disturbance over the last 100 years. Such a system will be in a steady state with respect to atmospherically derived ^{210}Pb and the flux (F) is related to the inventory (I) by the decay constant (λ) of ^{210}Pb as follows [73]:

$$F\,(Bq\,cm^{-2}\,yr^{-1}) \;=\; \lambda\,(yr^{-1}) \;\times\; I(Bq\,cm^{-2})$$

Values of the ^{210}Pb(atmos) flux calculated from soil measurements have been compared with flux measurements made from precipitation and dry deposition. Benninger et al. [8] give a flux value of 17 mBq cm^{-2} yr^{-1} for the eastern USA and flux estimates from soil profiles of between 13–20 mBq cm^{-2} yr^{-1}. The mean annual flux estimated from the inventory of ^{210}Pb in four soils in New Zealand (2.3–5.5 mBq cm^{-2} yr^{-1}) compared favourably with the flux measured in precipitation [2]. The global model of ^{210}Pb deposition developed by Turekian et al. [145] predicts an increase in the ^{210}Pb flux from west to east across the North American continent from $\sim$12 mBq cm^{-2} yr^{-1} on the west coast to $\sim$17 mBq cm^{-2} yr^{-1} on the east coast. However a recent review, by Urban et al. [149], suggests that the numerous reported measurements of atmospheric fluxes, by direct measurement (mean $\sim$17 mBq cm^{-2} yr^{-1}) and by measuring the soil inventory (mean $\sim$15 mBq cm^{-2} yr^{-1}) show a remarkable uniformity across most of North America. Interestingly the estimate of the mean ^{210}Pb flux from soil inventories is about 10% less than the value from direct measurement.

Graustein & Turekian [74] measured the ^{210}Pb inventories of forest soils at a range of altitudes in the eastern United States and found that the inventories at $\sim$1,000 m, or higher, are 2–3 times greater than the inventories in the lowlands. Using equations which predict the precipitation increase with altitude they concluded that their results could indicate an increase in ^{210}Pb concentration in rainfall of a factor of $\sim$1.5 at high altitude in comparison with rain falling on the lowlands. However, they were uncertain of the amount of cloudwater being intercepted by the trees. From the work of Lovett et al. [105] they estimated that occult deposition of cloudwater could introduce as much as up to 50% again of the

water falling as precipitation. If this additional input of water, on top of precipitation, were being deposited, an increased concentration of ^{210}Pb in precipitation is precluded.

2.1.12 Global and UK ^{210}Pb Fluxes

Typical global and UK ^{210}Pb fluxes are given in Table 2.3. Values tend to be smaller on to islands and where maritime airmasses predominate and larger values are found at more continental sites. The largest reported flux measurements are found for Japan which is immediately to the east, and thus downwind, of the Asian continent. Relatively few measurements of the ^{210}Pb flux have been made in the UK.

Reference	Location	Measurement	^{210}Pb(atmos) Flux ($mBq\,cm^{-2}\,yr^{-1}$)
Baranov & Vilenskii [3]	Moscow, Russia	direct	12
Baltakmens [2]	New Zealand	soil	2.3–5.5
Fukuda & Tsunogai [62]	Hokkaido, Japan	direct	37
Petit [128]	Belgium	pond	8.3–10
Glöbel et al. [68]	Saarland, Germany	direct	10
Peirson et al. [127]	Milford Haven, UK	direct	8.5
Clifton [35]	Plymouth, UK	direct	6.8
Eakins et al. [50]	Cumbria, UK	direct	15
Urban et al. [149]	USA	direct soil	17 15

Table 2.3: Measurements of the ^{210}Pb(atmos) flux to the ground for various locations on the globe

2.2 ^{137}Cs and ^{134}Cs in Air and Soils

The artificial radionuclide ^{137}Cs is produced in fission reactions via a gaseous precursor ^{137}Xe. It has only existed in the atmosphere since the advent of large-scale nuclear testing in the atmosphere in the early 1950's [73]. By far the most important source of ^{137}Cs in the atmosphere has been that produced in the atmospheric nuclear weapons tests, although significant releases have also occurred due to accidents involving nuclear reactors, for instance at Windscale in 1957 and Chernobyl in 1986, and at least one accident at a chemical and nuclear waste plant in the USSR in 1957, see for instance [30]. In addition to the ^{137}Cs deposited in global weapons fallout, some areas of the UK were subject to relatively large amounts of deposition from the Chernobyl reactor failure and the accident at Windscale in 1957 led to relatively high deposition within a few km of the plant but little at long range.

The fission yield of ^{134}Cs is very much lower than that of ^{137}Cs and the ^{134}Cs/^{137}Cs activity ratio in weapons fallout and reprocessing plant releases is of the order of 0.02. Furthermore, the relatively short half-life of ^{134}Cs (2.06 years) and the fact that large-scale atmospheric weapons testing ended in the early 1960's means that environmental inventories are generally near zero. However, the Chernobyl releases contained significant quantities of ^{134}Cs formed by activation, and the activity ratio (^{134}Cs/^{137}Cs) in rainfall after the accident was in the range 0.47–0.69 [23]. This characteristic ratio has since proved invaluable as a tracer of Chernobyl derived ^{137}Cs. Apart from differences in behaviour due to the different source terms for ^{134}Cs and ^{137}Cs, the two isotopes behave identically during transport and deposition.

2.2.1 Weapons Tests

The radionuclide ^{137}Cs has been present in the atmosphere in significant quantities only since the beginning of large-scale atmospheric testing of nuclear weapons in 1954. The intensity of weapons testing peaked between 1963 and 1964 and during

this period about half of the total injection of ^{137}Cs into the atmosphere took place [73]. In 1963 an agreement was signed by the USA, USSR and UK to end testing of nuclear weapons in the atmosphere. However, other countries did not sign and, China, France and India, for instance, have occasionally tested weapons in the atmosphere. It has been calcluated that up to 1978 about 960 PBq of ^{137}Cs had been injected into the atmosphere at a rate of 5.9 TBq per kT of explosive yield, [19][30]. Testing was carried out at altitudes ranging from a few metres above the ground up to about 100 km above the earth's surface [52].

The radioactive products of a nuclear explosion are separated into three fractions. The first is associated with large particles which fall to the ground in the vicinity of the test area on a timescale of a few hours, the second fraction is dispersed in the troposphere and is subject to the same scavenging processes as other atmospheric aerosols and deposits on a timescale of days, whilst the third fraction penetrates into the stratosphere and is deposited worldwide over a period of many months [52]. Before 1952, radioactive debris was restricted to the troposphere and the concentrations in air decreased at a rate corresponding to a residence time in the order of 7 days [23]. However, with the advent of high altitude and high yield (ie megaton) bomb testing, material was injected into the stratosphere, either directly or carried up in the convection currents associated with the fireball, and longer residence times were recorded. For tropospheric explosions generally, the residence time of radioactive debris was ~ 20 days. Eisenbud states that 'the principal source of worldwide contamination by long-lived radionuclides from bombs in the megaton range of yield is the component of the debris that was injected into the stratosphere' [52]. Stratospheric material is transferred to the troposphere during tropospheric folding events, a particularly common phenomenon during the winter and early spring in the northern hemisphere. The fraction of the radioactive debris which condenses out of the fireball and remains within the troposphere for more than a few hours and also the fraction returning to the troposphere from the stratosphere become associated with the ambient tropospheric aerosol and most activity is in the 0.3–1.0 μm size range [103].

2.2.2 Deposition of ^{137}Cs

Atmospheric ^{137}Cs is subject to the same processes of wet deposition and dry deposition as ^{210}Pb. Large particles with high dry deposition velocities are likely to deposit in the vicinity of the explosion; fission products from distant bomb tests become attached to 'natural' and pollutant derived condensation nuclei in the atmosphere of typical diameter 0.02–0.2 μm [29]. Wet deposition processes of rainout and washout become more important with distance from the source and this is the most effective removal mechanism for global fallout from the weapons tests [29]. The washout ratio is defined as the activity per kg of rain divided by the activity per kg of air (usually at ground level). A washout ratio value of a few hundred is typical for weapons fallout and Cambray et al. give a mean value of 730 for ^{137}Cs between 1960 and 1969 [22].

A team at Harwell exposed an artificial grass surface, consisting of strips of cellulose acetate, in the open air beneath a shelter. During periods in 1961 and 1962 when tropospheric nuclear tests were carried out in Russia the dry deposition velocities measured were of the order of 10 mm s^{-1}. For the period 1964–69, when activity was derived from the stratosphere, the mean dry deposition velocity to the artificial grass surface was 3.3 mm s^{-1} [30]. The dry deposition rate was observed to increase during periods of tropospheric fallout [126]. Cambray et al. concluded that dry deposition contributed about 17% of the total deposition of ^{137}Cs [22]. Other workers, however, have utilised smaller values of the dry deposition velocity in considering deposition of ^{137}Cs to the ground. Graustein & Turekian used a dry deposition velocity of 0.6 mm s^{-1} for ^{137}Cs when considering deposition to the lowlands of the eastern USA [73]. This amounted to less than 5% of the total deposition velocity (ie including dry and wet deposition) and they concluded that dry deposition is a minor pathway for the delivery of radionuclides to the lowlands of the eastern United States. Bunzl & Kracke used a value of 0.7 mm s^{-1} to estimate the dry deposition of ^{137}Cs to grassland sites in Germany [19].

A positive linear relationship, between the inventory of ^{137}Cs in soil and mean

annual rainfall, was first demonstrated by Peirson & Salmon [124], measurements were made at 7 UK sites. Cawse & Horrill also investigated the relationship between the ^{137}Cs inventory and mean annual rainfall, taking measurements from permanent grassland and woodland in England, Scotland and Wales [27]. Two sampling sites were chosen in each of 29 grid squares of 100 × 100 km at locations remote from nuclear installations. The mean accumulation of ^{137}Cs to 30 cm depth was observed to be 3,530 Bq m^{-2}, with average rainfall of 954 mm yr^{-1} at all soil sampling sites. This corresponded to a mean inventory of 3,700 Bq m^{-2} per 1,000 mm yr^{-1} rainfall, to 30 cm depth, in 1980 [14]. A similar relationship was found by Bunzl & Kracke for 25 grassland soils in Bavaria, Germany [19]. A correlation was also observed between ^{137}Cs deposition and mean annual rainfall for 25 forest soils (adjacent to the grassland sites). However, the results show 30% higher deposition on to forest soils than on to adjacent grassland. This finding was in contrast to that of Cawse & Horrill [27] who found no significant difference in ^{137}Cs deposition between grassland and woodland soils. The differences may be attributed to the altitude of the sites being sampled or to the coring location within the forest, where differences may be expected between the edge and points some distance into the forest. The forest sites sampled by Bunzl & Kracke may be receiving significant inputs due to occult deposition whereas the sites in the UK may not be. An important difference in the two studies is that, in Bavaria coniferous forest soil was sampled, whereas in the UK deciduous forest soil was sampled. At deciduous sites the interception of airborne fallout by woodland canopies will occur only during the vegetative growth period [19].

2.2.3 Chernobyl

Unit 4 of the RBMK–type reactor at Chernobyl, USSR exploded at 0123 hours on 26th April 1986 leading to a massive release of radioactivity to the atmosphere. A detailed account of the reactor design and the events leading up to the reactor failure and subsequent explosion is given by Eisenbud [52]. After the initial, and largest, release of radioactivity, major releases continued for about 10 days, virtu-

ally ceasing on 6^{th} May [30]. Cambray et al. estimate that, along with other radionuclides, a total of 70 PBq of ^{137}Cs (25% of the reactor inventory) were released to the atmosphere [23]. The initial release products were carried northwestwards towards Scandinavia, but from around 1600 hours on 26^{th} April the emissions were carried westwards, reaching the UK on 2^{nd} May 86 [140]. The cloud crossed the UK during the 2^{nd} and 3^{rd} and returned on $7/8^{th}$ May due to local recirculation. At Harwell the initial peak of activity in air ($0.5\,Bq\,m^{-3}$ of ^{137}Cs) was recorded on 2^{nd} May, followed by a much smaller peak on 7^{th} May and a broad peak in the second half of the month due to circulation of material eastwards round the northern hemisphere (in the order of $1\,mBq\,m^{-3}$). During June, the air concentration decreased with a mean atmospheric residence time of 7 days, similar to observations of low-yield weapons fallout [23]. Air concentrations remained higher than the levels attributable to weapons fallout throughout July, August and September over the UK, Cambray attributed this to continuing releases from Chernobyl until the reactor was finally sealed on 23^{rd} September. Continued high atmospheric concentrations at Munich-Neuherberg (Germany) were attributed to local resuspension of deposited material [84]. Most of the activity reaching western Europe was carried on particles of diameter less than $2\,\mu m$ [89].

Deposition of ^{137}Cs to the ground surface was very patchy over the UK. Clark & Smith have shown that the levels of deposited ^{137}Cs can be closely related to localised heavy rainfalls, during the passage of the cloud, which were associated with thunderstorms [33]. In particular, heavy rainfall fell over north Wales, Northern Ireland, Cumbria, southwest Scotland and the central Highlands of Scotland, whereas no rainfall was recorded over southern England during the 2^{nd} and 3^{rd} May. At Holmrook, Cumbria, a Chernobyl deposit of ^{137}Cs of $10{,}000\,Bq\,m^{-2}$ was measured, amounting to three times that due to weapons fallout, but at Orfordness deposition amounted to $110\,Bq\,m^{-2}$, only 10% of that due to weapons fallout. In some parts of northern and western Britain, for example areas of southwest Scotland, deposition exceeded $20{,}000\,Bq\,m^{-2}$ [33].

During the early phases washout ratios of the order of several thousands were

observed but towards the end of May a value of 410 was deduced [23]. From measurements made in the south of England, where no rain fell, a dry deposition velocity of around $0.5\,\mathrm{mm\,s^{-1}}$ was calculated, in line with estimates made by Chamberlain after the Windscale accident of 1957 [29][33]. Cambray et al. estimate that the Chernobyl accident increased the global inventory of ^{137}Cs by 5% and the UK inventory by 40% [23]. Clark & Smith estimate that $\leq 1\%$ of the total release of ^{137}Cs from Chernobyl was deposited onto the UK [33]. See Table 2.4 which lists the amount of ^{137}Cs deposited in the UK from the weapons tests, the Chernobyl accident and the 1957 Windscale accident.

Source	Activity (TBq)
Weapon testing	780
Chernobyl accident	320
Windscale accident	22
Total	1,122

Table 2.4: The UK inventory of ^{137}Cs, estimated from measurements in soil [23]. Tera (T) represents 10^{12}

2.2.4 ^{137}Cs in Soils

In temperate latitudes there is usually a cover of vegetation and Cs isotopes deposited from the atmosphere are likely first to make contact with vegetation surfaces. The quantities of Cs absorbed by plants in this way are extremely small although a significant proportion (10–40%) may be trapped on leaf surfaces [98]. Nevertheless, much of the deposit will be washed off onto the soil surface [157]. Fredriksson et al. make a distinction between mineral soils and organic soils in temperate lands [61]. They define mineral soils as soils containing appreciable, though varying, quantities of micaceous clay minerals and organic soils containing

little clay with a major component of organic matter at varying stages of decomposition. The binding of Cs by clays is attributed to either 'edge fixation' on ion exchange sites at the edges of interlayer spaces or 'interlayer fixation' which involves the diffusion of ions to interlayer sites where they are entrapped by collapse of the lattice [61]. Studies have shown that, in general, Cs is adsorbed irreversibly by micas and hydrobiotite, whilst montmorillonite and especially vermiculite hold Cs much less strongly, adsorption by 1:1 minerals such as kaolinite is very small [100]. Organic matter enhances the absorption of ^{137}Cs by plants, which implies that organic matter in soil inhibits the 'locking up' of Cs ions by clay minerals and leads to a larger fraction remaining readily exchangeable on mineral or organic particles and in solution (ie a greater fraction of the Cs ions remain 'labile'). This could be due to the organic matter in soil preventing contact between the clay minerals and Cs ions, or due to the sorption of organic molecules on clay surfaces interfering with the retention of Cs or a combination of the two [61]. Soil organic matter, as well as interfering with the adsorption of Cs by clays, sorbs Cs (possibly by ion exchange) and appears to act as a fairly slow release source [81]. It must be said, though, that the Cs-clay-organic system is very complicated and opinions differ. For instance, Cremers et al. belive that soil organic matter plays no part in the specific retention of Cs [39]. A range of factors determines the behaviour of Cs in soil, with respect to fixing and adsorption, but the most important factors are; clay mineral content and type, organic matter content, carbonate content and pH.

Downward migration of Cs occurs in soils either through downward transport in solution with the water flux (termed convection), or by diffusion, a process which is only important in dry soils. Migration down the profile is retarded by the fixing and adsorption processes outlined above, although Cs ions attached to mineral or organic particles may be transported downwards by particle migration if the particles are smaller than the soil pores [20]. Studies of the distribution with depth of ^{137}Cs from weapons testing in British soils has shown good retention in the top 20 or 30 cm or so of the profile for most soils.

Cawse & Horrill, in their survey of Britain, discovered that, on average, retention of ^{137}Cs in the 0-15 cm layer accounted for 75% of the amount recovered to 30 cm depth [27]. Cawse found that for a peat site in Sutherland, 83% of the total ^{137}Cs activity to 40 cm depth was present in the top 20 cm [26]. However, in comparison with a nearby mineral soil under permanent grassland, the recovery of ^{137}Cs to 30 cm depth in the peat was only half that in the soil and the accumulation in the soil was only about 60% of that expected from the annual rainfall to the site. Cawse considers that the low ^{137}Cs inventory measured in the peat may be an artefact of sampling peat with a narrow corer [26]. Bonnett et al. consider that where levels of ^{134}Cs and ^{137}Cs are lower than expected but there is no evidence of down-profile migration the shortfall may be due to considerable particulate radiocaesium erosion [12]. Studies in the Wye and Severn basins suggest that all Chernobyl-derived ^{137}Cs and about 90% of weapons fallout is contained in the top 5 cm of the soil profile. The soil types ranged from peat on hill tops, to podsols and gleys on the slopes with some brown earths and peat bogs in the valley bottom [12][13]. Livens et al. found (for 10 soils in upland Europe) that $>80\%$ of ^{137}Cs (and often 90%) is held in the top 10 cm of soil profiles [101]. Cawse and Horrill [27] found about 90% of ^{137}Cs in the fallout from weapons testing remained in the top 15 cm of 2 peat soils in the UK. Bunzl & Kracke [19] state that the rate of migration of ^{137}Cs is very slow and 95% of activity is still present in the 0–20 cm layers of most undisturbed soils. Bunzl [20] derived the migration rate of ^{137}Cs from the Chernobyl fallout and the global fallout of weapons testing in the horizons of a podzolic Parabrown forest soil under spruce and found that the average migration rates for Chernobyl ^{137}Cs are higher by a factor of 3–6 than those derived for the global fallout ^{137}Cs and concludes that Cs becomes less mobile in the soil as time increases. Livens & Baxter talk of a gradual 'locking up' of ^{137}Cs whereby the labile fraction of Cs decreases over time [99]. Howard et al. measured the ratio of ^{134}Cs / ^{137}Cs in a number of soils and in the NH_4^+ exchangeable fraction. Over time, the ratio in the exchangeable fraction fell from a near-Chernobyl value to more or less that of the whole soil.

Thus, the Chernobyl Cs was more easily exchanged shortly after the accident, but equilibriated with fallout over 1–2 years [85].

2.3 ^{7}Be in the Atmosphere

The natural radionuclide ^{7}Be, with a half-life of 53.3 days, is produced in the atmosphere by cosmic ray spallation of nitrogen and oxygen. The production rate of ^{7}Be displays a broad maximum at about 12 km altitude due to the downward decrease of the low energy neutron flux and the vertical distribution of air density. Production is greatest at the Poles, due to the deflection of cosmic rays by the Earth's magnetic field. However, tropospheric production rates are thought to be fairly independent of latitude. At low latitudes the maximum of ^{7}Be production occurs in the upper troposphere but shifts to the lower stratosphere above 30° N, primarily because of the decrease in the height of the tropopause [90]. Within the troposphere, production rates decrease by a factor of about 200 between the tropopause and ground level due to attenuation of the cosmic ray flux [76].

After production, ^{7}Be rapidly associates with ambient aerosol particles. In the stratosphere these particles are predominantly of diameter $\leq 0.1\,\mu$m and have a mean residence time of about 1 year above the tropopause. Since ^{7}Be has a mean-lifetime of about 77 days (the mean-lifetime is the average time that a radionuclide will exist after production and is given by the reciprocal of the decay constant, λ,) very little, under normal conditions, will enter the troposphere and the ^{7}Be flux to the ground will depend on the tropospheric production rate [43]. Only during injections of stratospheric air into the troposphere, for instance during folding events, is the ^{7}Be flux from the stratospheric reservoir significant [146]. In the troposphere, ^{7}Be is associated predominantly with sub-micron diameter aerosols. Measurements by Ludwick et al. showed 82% of ^{7}Be activity to be associated with aerosols $< 1.1\,\mu$m diameter [106].

Most authors have observed a seasonal variation of the ground level air concentrations of ^{7}Be, with the maximum concentrations typically a factor of between

2 and 5 times greater than the minimum, eg [76][125]. The maximum in ground level air concentration is most commonly observed in the late spring and is usually attributed to injection of stratospheric air or to a rise in the height of the tropopause, [43][83][120]. Feely et al. believe that the situation is more complex and attribute the seasonality in the ^{7}Be air concentration to a combination of four factors: injection of stratospheric air, vertical mixing within the troposphere (in mid latitudes), horizontal transport of air masses (affecting high latitudes) and the temporal pattern of precipitation (in areas subject to a seasonal precipitation pattern) [54]. The importance of each individual factor will vary according to the site location.

Table 2.5 gives some measurements of the ground level mean air concentration of ^{7}Be. Ground level concentration is usually a factor of at least 10 less than the tropospheric mean due to incomplete mixing [76]. The high value reported by Shapiro et al. was obtained at a relatively dry site subject to relatively infrequent precipitation events, so that ^{7}Be was allowed to build up in the lower troposphere [138]. Feely et al. give the results of measurements made at 28 stations located between 81° N and 90° S [54]. The lowest and highest reported mean concentrations are presented in Table 2.5.

Table 2.6 provides some measurements of the depositional flux of ^{7}Be to the ground. All authors point to precipitation scavenging (washout and rainout) as being the dominant depositional mechanism for ^{7}Be bearing aerosols. Olsen et al. [119] compared wet only and bulk samples and concluded that dry deposition accounted for less than 10% of total deposition; Todd et al. also quote a figure of about 10% for the dry deposition component [143]. Olsen et al. observed a high degree of correlation between the measured monthly deposition of ^{7}Be and ^{210}Pb despite their different sources and took this to reflect the importance of precipitation scavenging for both nuclides.

Typical average concentrations of ^{7}Be in rainfall are also given in Table 2.6 , with the assumption that all the deposition was by wet deposition. These values may thus be slight overestimates, due to the small fraction of ^{7}Be which is dry

deposited. The washout ratio, W, is defined as the activity per unit mass of rain divided by the activity per unit mass of air (usually at ground level) [29]. Todd et al. calculated an average value of 370 for the washout ratio for ^{7}Be in bulk deposition [143].

For ^{7}Be bearing aerosols, mean residence times in the range of 30–40 days are frequently quoted, for instance Shapiro et al. calculated a mean tropospheric residence time of 35.4 days [138]. In contrast, mean residence times calculated by observing the ratios of the radon daughters in air are generally smaller than this value, eg [30][50][73][111]. Graustein & Turekian calculated a mean residence time of about 5 days for ^{210}Pb- and ^{137}Cs-bearing aerosols and argued against a sharp increase in the mean residence time in the middle troposphere [73]. However, Shapiro et al. consider that the longer residence times calculated for ^{7}Be reflect its source in the upper troposphere, well above the cloud layer [138].

Inventories of ^{7}Be have been measured in marsh sediments and in soils [25][119]. Most of the ^{7}Be inventory generally occurs in the above ground vegetation (when present) and the values from soil and vegetated marsh cores compare favourably with inventories calculated from direct measurements. In unsaturated soils ^{7}Be may penetrate to depths as great as 10 cm, but where the groundwater level is frequently at or close to the marsh surface the total inventories of ^{7}Be and ^{210}Pb may be depleted in comparison with direct measurements of deposition [119].

An investigation of the atmospheric ^{210}Pb inventory with altitude, with measurements made at suitable sampling sites, gives information on the deposition pattern over the last $\sim$100 years. As most ^{210}Pb is deposited in rainfall, the inventory in soil with altitude will largely reflect the wet deposition pattern, integrated over many decades. In contrast, the short half-life of ^{7}Be (53.3 days) links measurements in the field to more recent rainfall events. The measured ^{7}Be inventory represents deposition over the past $\sim$270 days (ie 5 half-lives). There is a possibility to use the ^{7}Be inventory to investigate variations in the wet deposition pattern throughout the year. For instance, during the winter months rainfall is predominantly frontal in character, whereas, during the summer, convective

rainfall is common. There is also the potential to use ^{7}Be to investigate the precipitation scavenging pattern with altitude on an event basis. This approach may involve the use of artificial collectors, for instance muslin screens, or precipitation collectors. As the investigation of precipitation scavenging on a seasonal basis or on a single event basis was not a feature of this work the utility of the ^{7}Be data, from the Merrick, are limited in this study.

Reference	Location	Dates	Conc. of ^{7}Be (mBq m^{-3})
Crecelius [38]	Washington, USA, 46° N	Feb 76–Jan 77	4.20
Shapiro et al. [138]	California, USA, 34° N	Sep 73–Jul 75	8.00
Hötzl & Winkler [83]	Munich, Germany, 49° N	1983–85	3.63
Feely et al. [54]	Punta Arenas, Chile, 53° S	1970–85	1.22
Feely et al. [54]	Mauna Loa, Hawaii, 19° N	1970–85	7.47

Table 2.5: Mean concentration of ^{7}Be in ground level air at selected locations

Reference	Location	Dates	^{7}Be Flux (Bq m^{-2} d^{-1})	^{7}Be conc. in rain (Bq l^{-1})
Peirson [125]	Chilton, UK, 51° N	Oct 59–Sep 60	2.51	1.4
Peirson [125]	Milford Haven, UK, 51° N	Oct 59–Sep 60	2.37	1.4
Crecelius [38]	Washington, USA, 49° N	Feb 76–Jan 77	3.72	0.5
Turekian et al. [146]	Connecticut, USA, 41° N	Mar 77–Aug 78	10.4	2.7
Turekian et al. [146]	Bermuda, 33° N	Mar 77–Aug 78	7.78	1.7
Dibb [43]	Maryland, USA, 38° N	Mar 86–Nov 87	6.22	2.4

Table 2.6: Measurements of the flux to the ground and the concentration in rainfall of ^{7}Be at selected locations

Chapter 3

Experimental Methods

In this chapter the choice of mountains and the criteria for selecting individual soil sampling sites at each mountain will be discussed. A description of the field sampling procedure and sample processing is also included. The techniques for analysis of soil and vegetation samples by γ-ray spectroscopy, using the two Ge(Li) detectors at Edinburgh University are described together with the methods used in Loss-on-ignition measurements on samples taken at the Merrick and Ben Cruachan. Procedures for the estimation of the rainfall pattern at each mountain are also given.

In this study 'mountain' or 'mountain location' will refer to a mountain at which sampling was conducted. The term 'site' or 'sampling site' will refer to a patch of ground which was considered suitable, for the purposes of this study, for soil and vegetation sampling. A suitable site is one at which the inventories of ^{210}Pb, ^{137}Cs, ^{134}Cs and ^{7}Be in soil and vegetation are likely to reflect deposition from the atmosphere rather than any particular features of the site itself, for instance proximity to a river or the presence of burrowing animals. At many sampling sites, in order to define the intra-site variability, several sets of samples were taken. The term 'sampling point' or 'point' will refer to a position within a site at which a single pit or a bulked core was removed.

3.1 Sampling strategy and choice of mountain locations

The overall purpose of the experimental work was to establish the deposition pattern of ^{210}Pb(atmos), and to a lesser extent ^{137}Cs(bomb), on the western slopes of mountain locations. Western slopes were selected as, in the west of the UK, most of the rainfall originates from the west and southwest in frontal systems coming in from the Atlantic Ocean. The change in the deposition pattern was important, ie the magnitude of any increase or decrease with altitude, and the form of the deposition pattern in relation to the mountain profile was also of interest. The measured deposition pattern was compared to the pattern of average annual rainfall. This enabled inferences to be made about the rate of scavenging of aerosols with altitude and the importance of seeder-feeder scavenging, and occult deposition, mechanisms at mountain locations in northern and western Britain.

The initial objective was to sample a mountain of relatively simple topography, unobstructed from the western coast by intervening high ground and where there is a clear orographic enhancement of rainfall amount with altitude. The Merrick was chosen for the initial sampling expedition and work at this location gave an indication of the logistics necessary to carry out the project, the numbers of people necessary to assist in sampling expeditions, the timescale involved in sampling and analysis and thus the number of sites which could be sampled in the whole project. The next step was to sample at Great Dun Fell, the location with the best understanding of the processes controlling the deposition of acidic ions. Again, at Great Dun Fell, the topography is relatively simple and there is a large orographic enhancement of rainfall amount ($\times 1.7$ between the valley and the summit). The third stage of the project was to sample along a transect line, extending from the western coast inland for ~ 80 km, through the western Highlands of Scotland, the intention being to investigate how the deposition pattern with altitude varies with distance inland. Sampling was carried out at three mountains, Ben Cruachan, Beinn Dorain and Ben Lawers. Along this transect

line the orographic enhancement of rainfall amount declines from 3,600 mm yr^{-1} at Ben Cruachan to 2,500 mm yr^{-1} at Ben Lawers. Beinn Dorain and Ben Lawers are situated within the Highland mass and sampling at these two mountains gave data on the wet deposition field in areas of complex topography. See Table 3.1 which gives details of the 5 mountains and Figure 3.1 which shows their location in the UK. See also Fig 3.2 which shows the relief along the line of sampling for the Merrick, Great Dun Fell and the Highland transect. The line of sampling in each case has been extended westwards by at least 30 km to illustrate the topography 'upwind' of each mountain location. The details of the three transects are given in Table 3.2.

Mountain	Region	NGR	Height asl (m)	Sampling Dates
Merrick	Dumfries & Galloway	NX 42 85	843	26 Oct 88 & 30 Nov 90
Great Dun Fell	Cumbria	NY 71 32	847	19–26 Apr 89
Ben Cruachan	Strathclyde	NN 06 30	1,126	20 Jul 90
Beinn Dorain	Strathclyde	NN 32 37	1,074	16 Aug 90
Ben Lawers	Tayside	NN 63 41	1,214	9 Nov 90 & 10 May 91

Table 3.1: The five mountains selected for sampling in this study

At these five locations 66 individual sites were sampled, 8 at the Merrick, 23 at Great Dun Fell, 9 at Ben Cruachan, 11 at Beinn Dorain and 15 at Ben Lawers. The profile of each mountain and the position of the individual sampling sites are illustrated in Figures 3.3 and 3.4. From these sites a total of 519 samples of soil and vegetation were analysed. The following discussion will present the rationale behind the choice of each mountain.

Figure 3.1: The five mountain locations sampled in this study. Merrick (M), Great Dun Fell (GDF), Ben Cruachan (BC), Beinn Dorain (BD), Ben Lawers (BL)

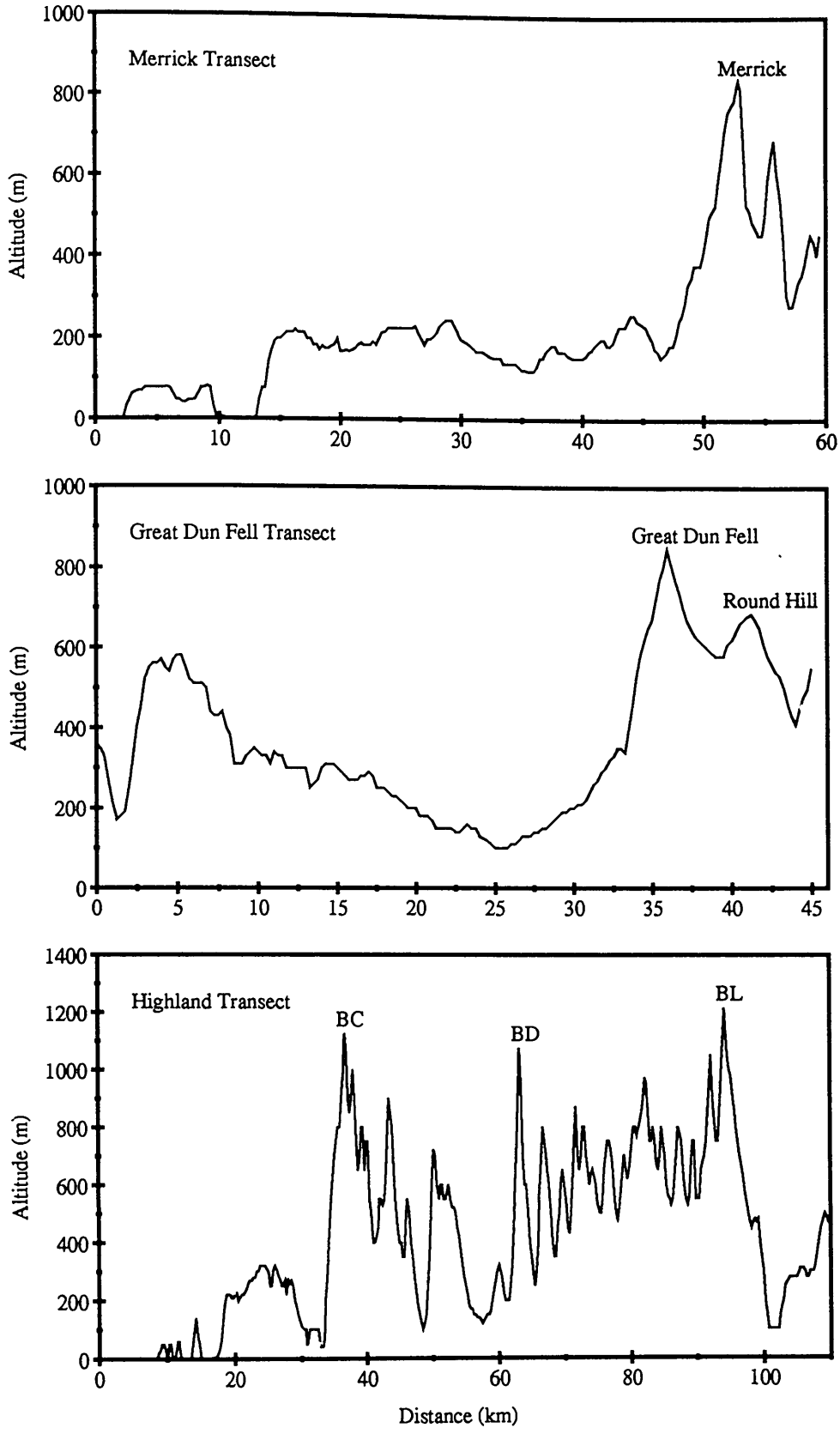

Figure 3.2: The relief at and to the west (ie upwind) of each mountain. Ben Cruachan, Beinn Dorain and Ben Lawers together form a transect through the west central Highlands

Transect	NGR Points	Bearing	Direction
Merrick	NW 944 638–NX 483 880	66°	WSW–ENE
Great Dun Fell	NY 480 040–NY 768 392	39°	SW–NE
Highland Transect:			
Ben Cruachan–Beinn Dorain	NM 715 200–NN 326 379	73°	WSW–ENE
Beinn Dorain–Ben Lawers	NN 326 379–NN 800 433	84°	W–E

Table 3.2: Details of the transect lines. The Highland transect changes direction by 11° at the summit of Beinn Dorain

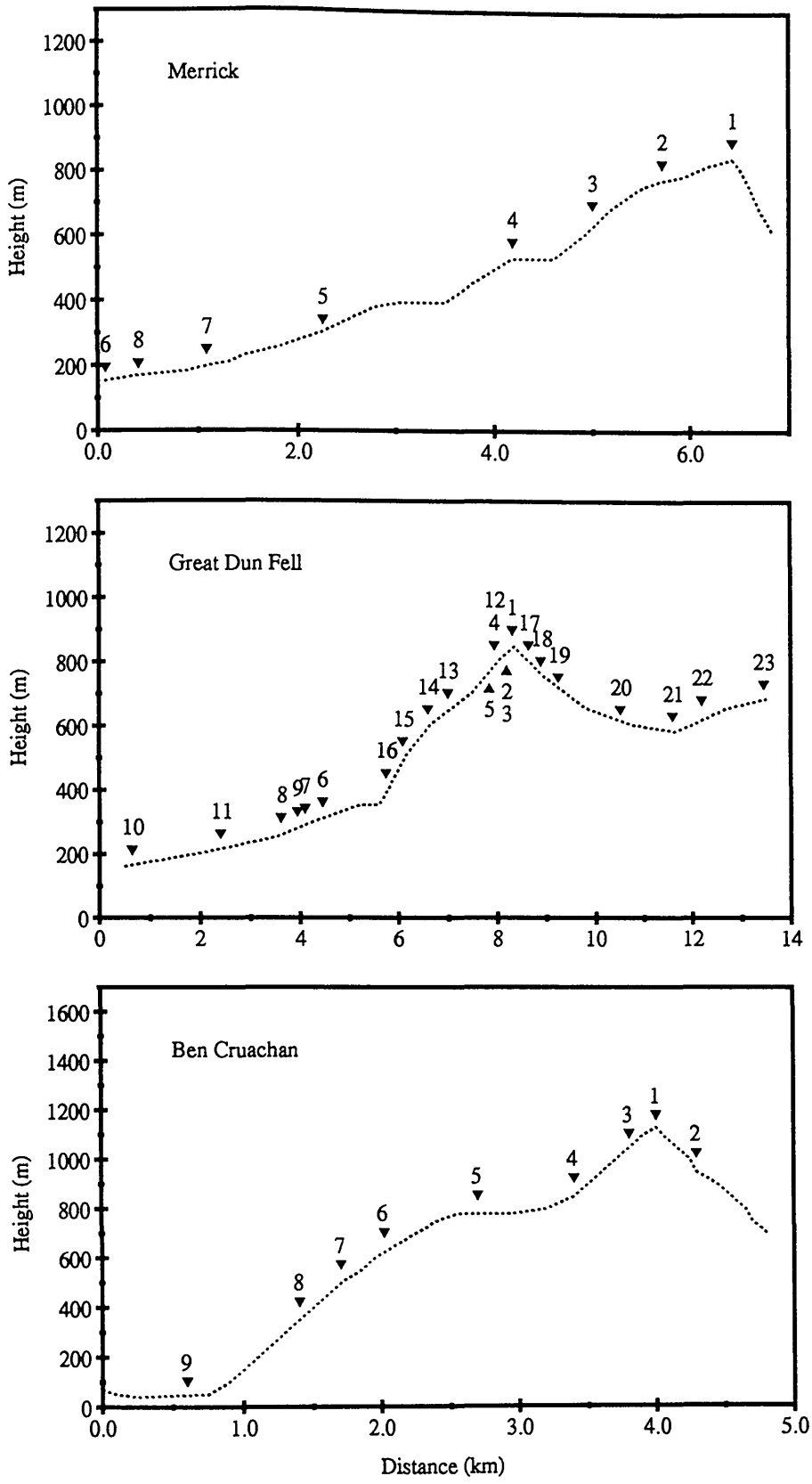

Figure 3.3: Hill profile and location of sampling sites for the Merrick, Great Dun Fell and Ben Cruachan

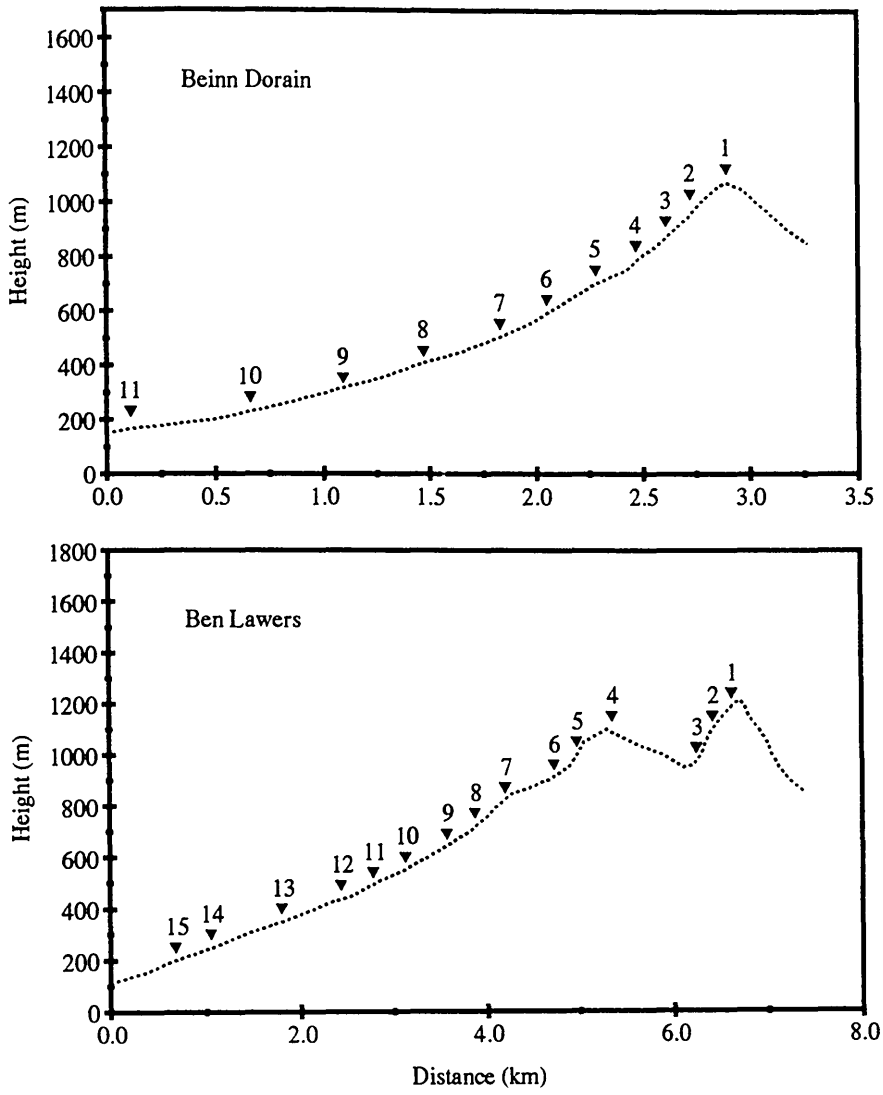

Figure 3.4: Hill profile and location of sampling sites for Beinn Dorain and Ben Lawers

3.1.1 Merrick

The Merrick was chosen as the first sampling location as it has a relatively simple topography (see Fig 3.3) and is subject to strong west and southwesterly airflow relatively undisturbed by hills upwind. However, even though there are no large hills, the Merrick transect (see Figure 3.2) shows that the mean height of the land from the base of the mountain to 30 km upwind is $\sim$200 m asl. Annual rainfall at the base of the Merrick (ie at sites M6 and M8) is about 1,800 mm yr^{-1}, annual rainfall at the western coast is $\sim$1,000 mm yr^{-1}. Thus, at the base of the mountain, rainfall amount is already enhanced to some degree.

The Merrick has features in common with Great Dun Fell, such as a similar altitude, simple topography and strong and relatively undisturbed west and southwesterly airflow. Cloud frequently shrouds the summit of Great Dun Fell and a similar situation is expected at the Merrick. Weston has developed a method for estimating cloud immersion frequency for locations in the UK based on cloud observations at meteorological stations and RAF stations [153]. This method estimates that the summit of the Merrick is shrouded in cloud for 30% of the year. As the Merrick has features in common with Great Dun Fell it was reasonable to expect that seeder-feeder scavenging and occult deposition mechanisms play a significant role in the scavenging of pollutant bearing aerosols at this location. An unbroken cover of peaty, highly organic soils, which strongly retain ^{210}Pb, enabled good samples to be collected, even at the summit. The Merrick presented itself as a good site to test the sampling and analysis technique and also to test the hypothesis that soil inventories of atmospheric ^{210}Pb may be used as an indicator of acidic deposition. An additional reason for selecting this mountain was that southwest Scotland was subject to high deposition of radioactive debris after the Chernobyl accident [33]. It was of interest to establish the pattern of deposition with altitude of ^{134}Cs and Chernobyl-derived ^{137}Cs at this location.

3.1.2 Great Dun Fell

Great Dun Fell is part of a ridge lying northwest–southeast with the tops of individual hills rising to ~ 900 m asl. To the southwest of the ridge lies the broad Eden valley, at ~ 150 m asl, see Fig 3.2. At the tops of the hills rainfall amounts are $> 2{,}000\,\mathrm{mm\,yr^{-1}}$ while in the valley the figure is $\sim 900\,\mathrm{mm\,yr^{-1}}$. This represents a considerable orographic enhancement of rainfall amount. It is known that the summit is in cloud for a part of 250 days per year [45]. The method of Weston [153] gives an estimate that the summit of Great Dun Fell is shrouded in cloud 28% of the time. Since the early 1980's a series of experiments have been carried out at Great Dun Fell to investigate the meteorology, microphysics, atmospheric chemistry and depositional characteristics of the site.

Fowler et al. [57] found that, on the western slopes of the hill when orographic cloud is present and a west or southwest wind is blowing the rainfall at the summit is on average twice that falling in the valley and that the concentrations of SO_4^{2-}, NO_3^-, Cl^-, NH_4^+ and H^+ in rainfall at the summit are $\sim \times 2.5$ those in rain falling in the valley. Thus, under certain conditions, wet deposition is a factor of 5 greater at the summit than in the valley. The observed enhancement in wet deposition was attributed to the seeder-feeder scavenging mechanism. Measurements at Great Dun Fell have been compared with the predictions of a model for the wet and occult deposition of sulphate over hills [32][80]. A clear qualitative agreement was found between the observations and model predictions showing an increase with altitude of the concentrations of SO_4^{2-} and NO_3^- in rainwater. The model of Hill et al. [80] also predicts the pattern of deposition of sulphate over the hill profile. Great Dun Fell is, thus, an important location for sampling as it allows a comparison of the measured inventory of atmospheric ^{210}Pb in soil with actual measurements of acidic deposition and the predictions of a model of sulphate deposition over hills.

The measurement of atmospheric ^{210}Pb in soil complements and extends the work carried out so far at Great Dun Fell. Fowler et al. present the results of

measurements made during 20 precipitation events from autumn 1984 and spring 1985 [57]. However, the atmospheric ^{210}Pb inventory in soil represents deposition over the last ~ 100 years. Assuming that on average there are in the region of 150 precipitation events per year, the inventory of atmospheric ^{210}Pb in soil will represent the atmospheric flux integrated over $\sim 15{,}000$ precipitation events. The soil inventory, thus, is a record of atmospheric deposition which has been collected over a much longer time-scale than measurements of acidic ions. Measurements were made by Fowler et al. at eight sites of different altitude on the western slopes of Great Dun Fell, whereas soil samples were collected at 23 sites, including the 'downwind' eastern slopes and at Round Hill, see Figure 3.3. Sampling at this number of sites on the mountain allows the pattern of deposition over the ridge to be investigated in detail, possibly identifying a point of maximum deposition and providing measurements to compare with the sulphate deposition model of Hill et al. [80]. The position of the maximum in the deposition profile of atmospheric ^{210}Pb, ie upwind of or downwind of or at the summit itself, will allow inferences to made on the occurrence and relative importance of aerosol scavenging mechanisms at this location.

3.1.3 Highland Transect

The aim of sampling along a transect line through the west central Highlands was to obtain data on the change in the soil inventory of ^{210}Pb(atmos) with distance inland from the western coast, and also on any change in the altitude dependence of the inventory with distance form the coast. Orographic enhancement of rainfall amount becomes less significant with distance from west to east across Scotland, see Figure 1.3. For instance, in the west, over the highest mountains, rainfall exceeds $3{,}600\,\mathrm{mm\,yr^{-1}}$, whereas in the east few areas experience rainfall of $> 1{,}200\,\mathrm{mm\,yr^{-1}}$. In order to produce maps for the wet deposition of acidic ions it is important to posess data on the change in the wet deposition field with distance inland from the western coast. The work carried out at the Merrick and at Great Dun Fell suggested that the soil inventory of atmospheric ^{210}Pb could be

used as an indicator for the deposition of acidic ions.

Sampling was conducted so as to gain information on how the importance of seeder-feeder scavenging and occult deposition changes with distance inland from the western coast. With this aim in mind, mountain locations were sought which would fulfill the following criteria; an altitude of $> 1{,}000$ m at the summit, a sufficient cover of soil for sampling purposes and a reasonably broad and deep valley 'upwind' to allow the airflow to descend to low level before encountering the main bulk of the mountain. The original intention was to assess the role and importance of seeder-feeder and occult deposition mechanisms at 'individual' mountains rather than at mountains at which the airflow is obstucted by upwind topography. However, in the event, this proved more difficult than was anticipated. Three mountain locations were selected for sampling, Ben Cruachan, Beinn Dorain and Ben Lawers, situated respectively 30, 55 amd 80 km from the western coast. Beinn Dorain is situated 26 km east-northeast of Ben Cruachan and Ben Lawers is 31 km east of Beinn Dorain. Figure 3.5 shows the average annual rainfall along the transect line. Rainfall amount is seen to decrease from west to east across the highland massif, areas of maximum rainfall are associated with mountain peaks. It is important to note that rainfall is less at Ben Lawers than at Ben Cruachan even though Ben Lawers is the highest mountain.

Ben Cruachan was selected for sampling because of its proximity to the coast and because of the very large orographic enhancement of rainfall amount experienced at this location. Average annual rainfall near to the summit is 3,600 mm yr^{-1} and this mountain is among the wettest places in Britain. The Pass of Branden lies southwest of the mountain and airflows from the southwest quadrant are relatively unobstructed by high ground upwind. It must be said, though, that the island of Mull (rising to 966 m) lies directly to the west and the islands of Jura (rising to 784 m) and Islay lie directly to the southwest. The southwest face of the mountain is very steep with an average gradient of 1 / 3.6, thus strong updrafts may be expected at this location.

Beinn Dorain was selected as the best location downwind of Ben Cruachan.

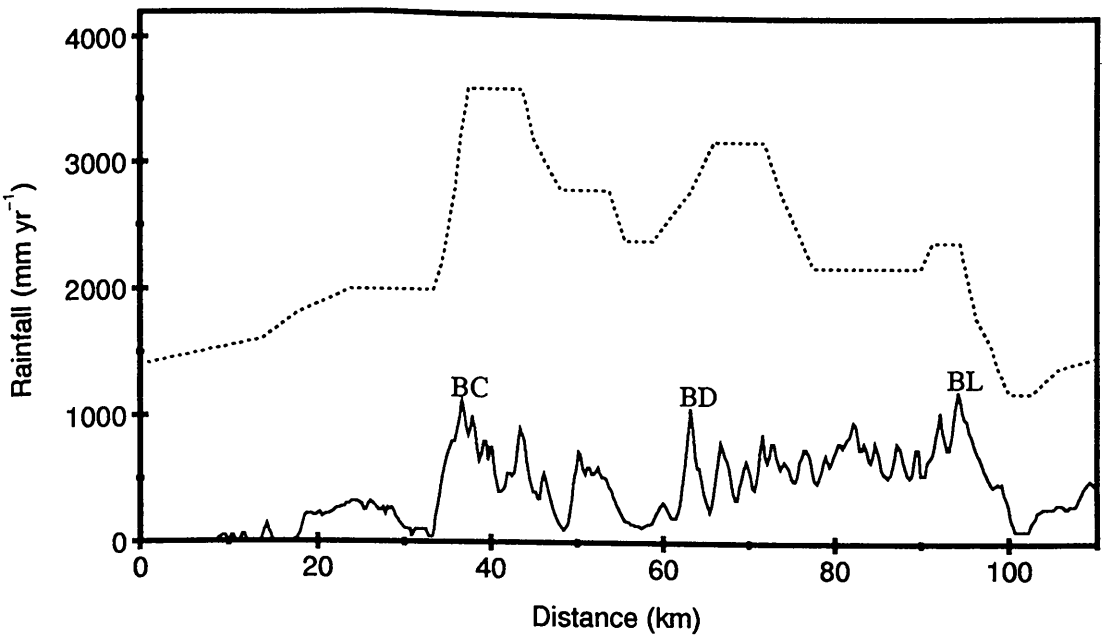

Figure 3.5: Average annual rainfall along the Highland transect (dotted line). The solid line represents the topography along the transect

The valley of the Allt Kinglass, lying to the west and southwest of the mountain, allowed sampling to be conducted over a large altitude range, ie 210–1,074 m. However, it is uncertain whether the valley is wide enough to allow airflows from the southwest quadrant to descend to low level before encountering the mountain. There may be a possible 'funneling' of airflows by Glen Orchy which runs southwest to northeast and lies at the base of Beinn Dorain. The southwest face of this mountain is even steeper than that of Ben Cruachan with an average gradient of 1 / 2.5. Sampling was facilitated at this location by an unbroken covering of soil, even at the summit.

Ben Lawers was chosen as the third sampling location of the transect. Situated 80 km from the coast it rises to an altitude of 1,214 m. The presence of Loch Tay to the south and southwest of the mountain enabled samples to be taken from a large altitude range, ie 200–1,190 m. The topography of the mountain to the southwest is relatively simple (less steep than Ben Cruachan and Beinn Dorain) and a path running between the visitor centre and the summit helped in the

sampling expeditions. Soil cover at this mountain was complete except around the summit itself. Airflow from a southerly or southwesterly direction is able to descend to low altitude before encountering Ben Lawers. However, as can be seen in Fig 3.2, directly to the west Ben Lawers is obstructed by high ground which has a mean height of $>600\,\mathrm{m}$. Ben Lawers is almost equidistant from the west and east coasts, being situated about 110 km from the east coast. The calculation of Weston [153] predicts that the summits of Ben Cruachan, Beinn Dorain and Ben Lawers will be immersed in cloud for 39, 37 and 40%, respectively, of the time.

3.2 Field Sampling

3.2.1 Selection of sites

Care was taken in selecting individual sampling sites. The object was to sample sites which hold an inventory of radionuclides representative of atmospheric deposition at their elevations. In order that the soil inventory of atmospheric ^{210}Pb be in steady state with the mean depositional flux from the atmosphere the soil must have been free from disturbance for ~ 100 years (ie ~ 5 half-lives of ^{210}Pb). Sites were avoided at which there was a possibility of overland flow and soil erosion, for instance on steep slopes, and at which deposition of soil particles was a possibility, ie sites on river flood plains. Areas prone to large animal burrowing and where recent ploughing may have occurred were also discounted for sampling. A detailed investigation of the interaction between forest and atmospheric deposition mechanisms was not within the scope of this study and thus woodland and shrubland were avoided. However, a single woodland site, G8, was sampled at Great Dun Fell so as to compare with neighbouring, non-woodland, sites. Narrow valleys, in which the airflow may be complex, were also avoided.

Sites were sought on land exposed to the west or to the south at which the ground was level or gently sloping, far from trees or bushes and where the soil was not too stoney. However, at some elevations, sampling on steep gradients

was unavoidable, for example sites D5 and D6 at Beinn Dorain. At the higher elevations there was very little chance of sampling sites having been disturbed recently by human activity, but at lower levels, in the valleys, suitable sampling sites were more difficult to find. Where possible, local farmers were consulted on land-use during the present century.

It must be noted that the cross-sectional area of the soil samples was extremely small in comparison to the area of the mountains, the diameter of the small corer being 6.4 cm and that of the large corer 10.2 cm. In order to compensate for the fact that individual cores represented a very small area, several cores were collected from each site. This gave an indication of the intra-site variability of the soil inventory for each radionuclide. From most sampling sites, if using the small corer, 5–15 cores were collected and, if using the large corer, 2–6 cores were collected.

Except at Beinn Dorain, sampling at each mountain took place along a line running in a roughly southwest to northeast direction. Suitable sampling sites were sought as close as possible to this line. At Beinn Dorain it was not possible to sample along a southwest–northeast transect line due to the very steep gradient of the southwest face. Instead, the line of sampling proceeded due north from the summit and then due west, towards the Bridge of Orchy.

3.2.2 Field sampling procedure

Following selection of a suitable site, the position and elevation, measured with an altimeter, of the site were recorded and any outstanding features of the site were noted. Vegetation samples were taken with garden shears from an area of 1 m^2 cut down to 1 cm from the ground surface. Pit samples were collected using a flat spade of width 20 cm producing a pit area of 400 cm^2. All the soil was removed from the pit in 5 cm depth intervals.

Most of the field samples were taken with soil corers. In all, three types were used. On the first sampling expedition to the Merrick a small corer supplied by ITE Merlewood was employed; having a diameter of 7 cm and could collect cores

Figure 3.6: The large corer (10.3 cm diameter) with two soil cores excavated at Beinn Dorain

up to 10 cm in length. All subsequent soil samples were taken with either a small brass corer of diameter 6.4 cm and length 16 cm or a large corer adapted from a plastic drainpipe of diameter 10.3 cm and length 30 cm. The large corer was ideal for use on spongy, highly organic soils and the small brass corer was used for sampling more resistant mineral soils. See Figure 3.6 which shows the large corer with two soil cores, which have just been excavated.

Normal procedure at a sampling point was to take a number of cores, 5 if using the small corer and 2 if using the large corer. With a sharp knife each individual core was subdivided from the top downwards into sections, measured with a ruler, representing depth intervals. The top section cut comprised the vegetation, including undecayed organic matter, and the first 1 cm of soil. The equivalent depth intervals from each of the five, or two, cores were bulked to form one sample. Thus each individual sample from a sampling point represented material from more than one core.

At the Merrick (sites M1–M6) and at some sites at Great Dun Fell cores were

subdivided into 0–1 cm and 1–10 cm sections. However, it was decided that greater resolution with depth down the core was desirable as there was a danger of diluting the ^{210}Pb(atmos) which is predominantly found in the top few centimetres of the soil. Subsequently, cores were subdivided into 0–1, 1–5 and 5–10 cm sections (and at 5 cm intervals down to the bottom of the core). At some sites the 5–10 cm section was further subdivided into 5–7.5 cm and 7.5–10 cm sections.

Although soil profiles were recorded at some sites, a detailed consideration of the soils encountered did not constitute a part of this study. A similar pattern of changing soil type with altitude was observed at the five mountain locations. On the mountain slopes, between $\sim$300 to $\sim$800 m asl, soils are highly organic in character, for instance blanket peat soils and peaty gleyed podsols. Below $\sim$300 m, soils are more mineral in character, for instance acid brown soils and related gley soils. At the highest elevations, especially at Ben Cruachan, Beinn Dorain and Ben Lawers above $\sim$800 m, thin humus soils are found with bare rock exposed in places [156]. Most of the summit area at Ben Cruachan was exposed rock.

In summary, most samples were taken using soil corers, either the large corer which could take samples down to 20 cm depth, or the small corer which could take samples down to 15 cm depth. Cores were subdivided and bulked togther at the site. At many sampling sites bulked core samples were collected from several points (usually 3 points) to examine the intra-site variability in the radionuclide inventories.

3.2.3 Sample processing

All individual soil and vegetation samples were dried overnight at 80°C. The samples were weighed before and after drying. Almost all samples were processed prior to γ-ray analysis using the large ovens, mills and rotary sieve (2 mm mesh size) at ITE Merlewood. The core samples taken from the Merrick on 26th Oct 1988 were, however, processed at Edinburgh University. Vegetation samples and peat soil samples with a low stone content were ground using a knife-mill. Stony

soil samples were sieved to remove stones above 2 mm in diameter. Any residue of stones above 2 mm diameter was discarded and the remaining soil sample re-weighed. The material > 2 mm diameter was discarded on the assumption that they are closed systems which do not contain any atmospherically derived Cs and in which ^{226}Ra and ^{210}Pb are in radioactive equilibrium. Stones would thus only serve to dilute the counted sample [73].

3.3 Sampling of each mountain

3.3.1 Merrick

Initial sampling was carried out at the Merrick on 26^{th} Oct 1988, soil and vegetation samples were taken from 6 elevations (sites M1–M6) between the valley containing the Water of Minnoch to the west and the summit, see Table 3.3. At the lowest four sites (M3–M6) three vegetation samples were taken at each site, but there was insufficient vegetation at the top two sites (M1 and M2). At each site a single pit was dug and soil taken from 0–5, 5–10, 10–20 and 20+ cm depth intervals. In addition, bulked cores were taken from 3 points at each of the 6 sites and subdivided into 0–1 and 1–10 cm sections.

To give an indication of the scale of the work involved in collecting samples; on the first Merrick sampling expedition (26^{th} Oct 1988) six people were involved collecting and transporting 40 kg of soil from six sites (M1–M6), along a distance of 7 km, during one day. At Great Dun Fell, during the afternoon of 26^{th} April 1989, six people helped collect and remove 46 kg of soil from seven sampling sites (G17–G23) along a distance of 6 km. The sampling situation was similar at the other three mountains. The number of points which could be sampled, during one day, depended on the distance to be covered and the fact that each person could carry no more than about 8 kg of soil. Mountain roads, for instance on the west of Great Dun Fell, greatly facilitated the sampling.

The bulked core samples taken on the first expedition were ground using a

pestle and mortar in the laboratory at Edinburgh University. Any stones were removed although these were not accounted for by re-weighing the sample. This could lead to an overestimate of the ^{210}Pb(atmos) content of the core samples. However, data from the sieved pit samples, for which the >2 mm material was weighed, suggests that the overestimate, in the values from the core samples, is no greater than 10% at any site, and is thus considered to be insignificant. These bulked core samples from the Merrick were the only ones not to be processed at ITE Merlewood.

In order to provide further information on the radionuclide inventories at low altitude, additional sampling was carried out on 30th Nov 1990. Two bulked cores were collected from 198 m and two from 164 m altitude (sampling sites M7 and M8 respectively) (see Table 3.3). Care was taken to ensure that these sites lay on the same transect line as the initial six sites.

Site	Altitude (m)	NGR Square NX	No. of points	Vegetation samples	Sample depth (cm)	Notes
M1	843	428 855	4	–	20	
M2	777	426 852	4	–	25	
M3	648	412 837	4	3	20	
M4	530	405 832	4	3	20	
M5	290	386 839	4	3	20	
M6	150	371 829	4	3	25	near river
M7	198	381 835	2	–	15	
M8	164	374 831	2	–	20	wet, near bog

Table 3.3: Details of each sampling site at the Merrick. Sites M1–M6 were sampled on 26th Oct 1988, sites M7 and M8 on 30th Nov 1990

3.3.2 Great Dun Fell

Sampling took place on Great Dun Fell between the 19^{th} and 26^{th} April 1989, roughly along a transect line running in a southwest to northeast direction (NY 658 257 to NY 743 362). On the southwest facing slopes of Great Dun Fell between the Eden Valley and the summit 16 sites were sampled (G1–G16) and northeast of Great Dun Fell between the summit and Round Hill 7 sites were sampled (G17–G23). Table 3.4 lists the sites sampled. It was difficult to identify suitable low altitude valley sites at which there was little chance of recent disturbance, but eventually 6 sites were found (G6–G11). One of these sites (G8) was a small patch of woodland and so the values from this site will be treated with care in the analysis. All the sites sampled at Great Dun Fell had at least 10 cm of soil cover. At most sites core samples were taken down to at least 15 cm depth.

3.3.3 Ben Cruachan

Sampling took place at Ben Cruachan on 20^{th} July 1990 along a transect line running in a west-southwest to east-northeast direction (from NN 032 290 to NN 083 310). Samples were taken from eight different elevations between the valley of the River Awe (to the southwest of Ben Cruachan) and the summit. An additional sample was taken at a site on a col due east of the summit (site C2). See Table 3.5. Due to pressure of time and limited personnel, only one bulked core was taken from each site except C5 at 750 m altitude where two bulked cores were taken. This site was on a level piece of ground below a steep slope. The lowest lying site, C9, was situated near to the River Awe and the results suggest that this site has suffered a disturbance of some nature recently.

3.3.4 Beinn Dorain

Beinn Dorain was sampled on the 16^{th} Aug 1990. Bulked cores were taken from 11 different elevations between the summit and the valley of the Allt Kinglass to the west. Because of the extremely steep slope of Beinn Dorain to the southwest

Site	Altitude (m)	NGR Square NY	No. of points	Sample depth (cm)	Notes
G1	840	711 322	3	15	
G2	830	711 320	3	10	
G3	820	711 319	1	15	
G4	790	711 317	3	15	
G5	770	714 316	3	15	
G6	310	686 289	3	15	
G7	290	684 290	1	15	
G8	260	681 284	3	15	copse
G9	280	683 285	3	15	
G10	160	686 255	3	15	near river
G11	210	698 247	3	15	above stream
G12	800	709 320	3	10	
G13	650	702 313	3	10	
G14	600	701 308	3	10	
G15	500	694 307	3	10	
G16	400	692 305	3	15	
G17	800	711 325	3	10	
G18	750	713 328	3	10	
G19	700	715 330	3	20	
G20	610	728 339	2	10	
G21	580	728 346	3	15	above stream
G22	630	735 354	3	10	
G23	680	744 361	3	20	

Table 3.4: Details of the sampling sites at Great Dun Fell. G8 is a woodland site. Sites G1–5 were collected on 19^{th} Apr 1989, sites G6–8 on 20^{th}, site G9 on 21^{st}, sites G10–11 on 25^{th} and sites G12–23 on 26^{th}

Site	Altitude (m)	NGR Square NN	No. of points	Sample depth (cm)	Notes
C1	1,126	070 305	1	15	narrow ledge
C2	950	077 304	1	15	
C3	1,050	066 305	1	15	'bowl' in rock
C4	850	059 302	1	15	
C5	780	057 299	2	15	
C6	630	052 301	1	20	
C7	500	047 299	1	20	
C8	350	045 295	1	20	
C9	50	039 292	1	10	near river

Table 3.5: Details of the sampling sites at Ben Cruachan. Sample collection took place on 20^{th} July 1990

sampling along a southwest–northeast transect line was not possible. The sites D1 to D4 are situated on a north–south line (NN 325 370 to NN 325 400) from the summit and along the top of a ridge running northwards to a corrie, Coir an Dothaidh. The sites D5 to D11 lie along a west–east line (NN 300 395 to NN 330 395) running from close to the corrie, Coire an Dothaidh, to the Bridge of Orchy station. See Table 3.6. Figure 3.7 shows the profile of Beinn Dorain from a southerly viewpoint, the steepness of the southwest face can be clearly seen. Figure 3.8 shows low level, orographic cloud at this location on the day of sampling.

3.3.5 Ben Lawers

Sampling of Ben Lawers took place in two stages, the first on 9^{th} Nov 1990 when sample were taken from 12 elevations between the visitor centre (NN 6115 380) and the summit of Ben Lawers (NN 636 414). The transect line running southwest–

Site	Altitude (m)	NGR Square NN	No. of points	Sample depth (cm)	Notes
D1	1,074	326 379	3	10	
D2	980	323 388	1	10	
D3	880	323 394	1	10	
D4	790	325 396	3	10	
D5	700	320 395	1	10	very steep
D6	590	317 395	1	10	very steep
D7	500	315 394	3	15	
D8	400	311 394	1	20	
D9	300	307 394	1	20	
D10	250	305 394	1	15	
D11	210	302 394	3	15	

Table 3.6: Details of the sampling sites at Beinn Dorain. Sample collection took place on 16^{th} Aug 1990

Figure 3.7: Beinn Dorain as seen from the south

Figure 3.8: The base of a cap cloud shrouding the upper reaches of Beinn Dorain on the day of sampling

northeast followed the path leading from the visitor centre over the summit of Beinn Ghlass (1,100 m asl) along a col and up to the summit of Ben Lawers.

The second phase of sampling took place on 10^{th} May 1991, four sites were sampled between the visitor centre and Loch Tay along a line running north–south (NN 615 380 to NN 615 350). There is little or no evidence of recent disturbance at these four low altitude sites. Three points were sampled at each site to give good precision on the, low altitude, baseline values. The individual sampling sites are listed in Table 3.7. At site L12, close to the visitor centre, one point was sampled during the first expedition and three points were sampled during the second expedition.

Site	Altitude (m)	NGR Square NN	No. of points	Sample depth (cm)
L1	1,190	635 415	1	10
L2	1,100	633 414	1	15
L3	975	630 412	1	15
L4	1,100	626 405	1	10
L5	1,000	623 402	1	15
L6	910	622 399	1	15
L7	820	621 395	1	15
L8	720	619 393	2	15
L9	640	615 394	1	20
L10	550	614 387	3	20
L11	490	613 383	1	15
L12	430	612 380	4	30
L13	350	615 375	3	15
L14	250	617 368	3	15
L15	200	610 360	3	15

Table 3.7: Details of the sampling sites at Ben Lawers. Samples were collected from sites L1–L12 on 9^{th} Nov 1990 and from sites L12–L15 on 10^{th} May 1991

3.4 Gamma-ray Analysis

When an atomic nucleus undergoes radioactive decay the daughter product is often left in an excited energy state and may reach its ground state through the emission of γ-rays. For example, a ^{137}Cs nucleus changes into ^{137m}Ba through beta-emission. The resulting ^{137m}Ba nucleus de-excites, to its ground state, through the emission of a 661.7 keV γ-ray. The proportion of ^{137}Cs decays which lead to a 661.7 keV γ-ray being produced is known as the branching ratio, b_γ. The 661.7 keV energy is indicative of ^{137}Cs decay and measurement of the count-rate of 661.7 keV photons will allow determination of the number of ^{137}Cs nuclei present in the analysed sample. Other radionuclides produce γ-rays of characteristic energy, see Table 3.8, and measurement of the count-rate allows their concentration in a sample to be determined.

The following sections will describe the main interaction processes of γ-rays with matter, their detection using Ge(Li) detectors and determination of the concentration of specific radionuclides in a sample.

3.4.1 Interaction of Gamma-rays

There are three major interaction processes of γ-rays with matter, photoelectric interaction, Compton scattering and pair production [53]. In a photoelectric interaction the γ-ray photon interacts with an absorber atom and completely disappears. The energy of the photon is transferred to one of the atomic electrons which is ejected from the atom. This photoelectron posesses a kinetic energy given by:

$$KE_e = h\nu - E_b$$

where E_b represents the binding energy of the photoelectron and $h\nu$ represents the energy of the absorbed photon. The absorber atom de-excites through the emission of one or more characteristic X-rays or the emission of an Auger electron. Any emitted X-rays are subsequently photoabsorbed and the result of a photoelectric interaction is that all the energy of the γ-ray is transferred into kinetic energy

of electrons. This mode of interaction is ideal for detection since a photoelectric interaction in the sensitive region of the detector will result in all the energy of the γ-ray being deposited. In Compton scattering an incoming γ-ray photon collides with a relatively unbound electron in the absorbing material and is deflected from its original path. Only a proportion of the γ-ray energy is transferred to the electron. It is possible for an incoming γ-ray to undergo Compton scattering, and thus tranfer a fraction of its energy, and then undergo photoelectric absorption with the end result that all the energy of the photon is absorbed. Pair production is only possible for γ-rays of energy greater than 1.02 Mev [93].

3.4.2 Ge(Li) Detectors

To determine the radionuclide content of the collected samples two Ge(Li) detectors belonging to the Edinburgh Nuclear Physics Group were used. A Ge(Li) detector consists of a germanium crystal which has been ion drifted with mobile lithium ions. This process results in a relatively thick active volume (ie a few mm) which is important in detecting γ-rays. The crystal has a reverse biased n-i-p configuration and the high resistivity intrinsic zone, i, is the sensitive region. Typical bias voltages are 500–4,000 V across intrinsic regions of 5–10 mm thickness [93]. A γ-ray photon entering the sensitive region may interact and transfer some or all of its energy to the electrons in the crystal. These excited electrons will cause further ionization of atoms in the crystal, producing many electron-hole pairs which are swept out of the active region by the applied electric field. The lifetime of the charge carriers (electrons and holes) is much greater than the time required to collect them at the boundaries of the intrinsic region. This current of electrons and holes forms the basic signal information from the detector [93].

The number of electron hole pairs formed in the active region is proportional to the amount of energy deposited. The energy required to create an electron hole pair in Ge(Li) detectors is about 3 eV [93], and thus a single γ-ray photon will produce many charge carriers, giving good statistics in the measurement of the current pulse from the detector. As a result Ge(Li) detectors have very good

energy resolution compared to other γ-ray detectors [113]. Other factors which limit the energy resolution of the detector are variations in the charge collection efficiency and the broadening effects due to the contributions of electronic noise [93].

After the current pulse from the pn diode has passed through a pre-amplifier it is taken to a shaping amplifier which produces an output pulse of height proportional to the energy deposited in the active region of the detector. The signal from the shaping amplifier is then passed into a Multi Channel Analyser (MCA) which records the output pulse-height spectrum. The horizontal axis of the spectrum consists of a large number of channels, typically a few thousand, each of which corresponds to a slightly different energy and the vertical axis represents the number of counts in each channel. The MCA measures the height of each pulse output from the shaping amplifier and adds a count to the relevant channel. Using γ-rays of known energy the pulse-height spectrum may be calibrated and will show the relative occurrence of different energy depositions in the sensitive region of the detector.

A γ-ray photon which undergoes a photoelectric interaction in the sensitive region of the crystal will deposit all its energy and will result in a count in the full energy peak (FEP) in the pulse height spectrum. A γ-ray photon which is Compton scattered in the sensitive region and deposits only a proportion of its energy will produce a count in the region of the spectum lower in energy than the full energy peak, in the region called the Compton continuum. It is possible, though, that the γ-ray photon will deposit all its energy in the active region through a combination of Compton and photoelectric interactions and thus produce a count in the full energy peak of the spectrum. Figure 3.9 shows a spectrum obtained with a ^{60}Co source and a Ge(Li) detector. The full-energy peaks of the 1,173 and 1,332 keV γ-rays emitted by the source are shown along with the lower energy continuum caused by Compton interactions with electrons in the sensitive region of the detector. The peaks have been cut so as to show the form of the Compton continuum.

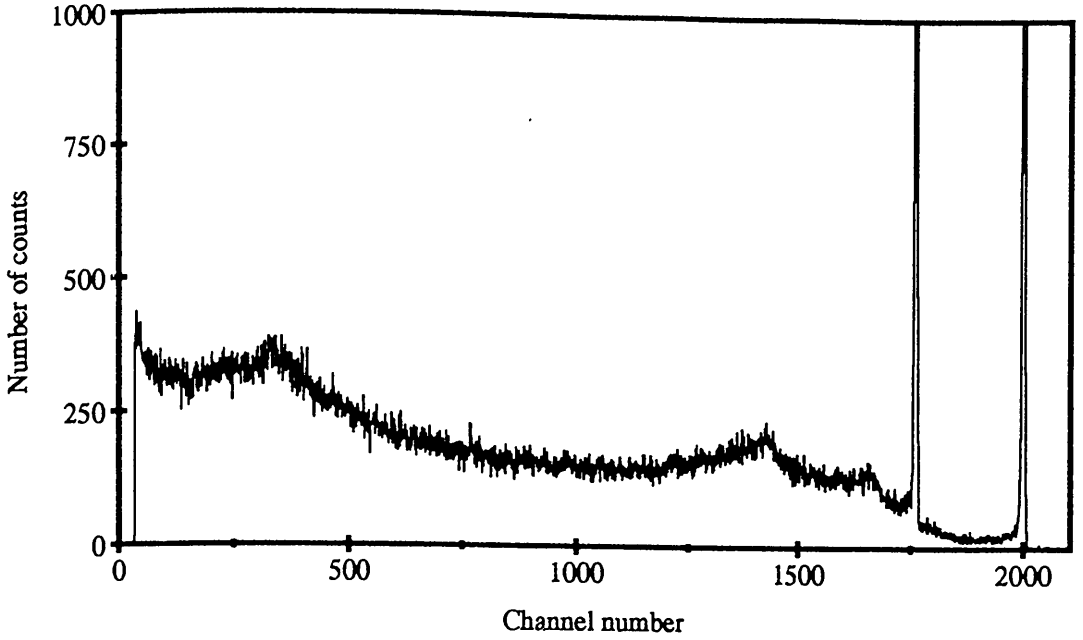

Figure 3.9: A γ-ray spectrum obtained using a ^{60}Co source showing the full-energy peaks of the 1,173 and 1,332 keV γ-rays and the lower energy Compton continuum. The two peaks, respectively, rise to 3,360 and 2,938 counts per channel

The concentration of specific radionuclides in the analysed samples was determined by summing the number of counts in the individual channels which make up the full energy peak and dividing by the length of time that the sample was under analysis. This gives a count rate for that radionuclide incident on the detector. Each collected sample was analysed for a variety of radionuclides and the excellent energy resolution of the Ge(Li) detectors allowed the concentrations of the different radionuclides to be measured from a single spectrum. The radionuclides of interest in this study and the energies of their characteristic γ-rays are shown in Table 3.8.

3.4.3 Anti-Compton Shield

One of the two Ge(Li) detectors used in the γ-ray analysis of the samples, a 19% relative efficiency crystal, was fitted with an anti-Compton shield. The Ge(Li) is surrounded by an annular NaI(Tl) detector which is set up to run in anti-coincidence mode with the Ge(Li). If a γ-ray photon undergoes Compton scatter-

ing within the Ge(Li) detector and then exits and also interacts in the surrounding NaI(Tl) crystal the signal from the NaI(Tl) will lead to the signal from the Ge(Li) being rejected. This potential count in the spectrum is now rejected. Counts representing cosmic rays, which are 99% relativistic muons, are also rejected. To get to the Ge(Li) they would have to pass through the anti-coincidence shield and hence would leave a signal. Running the 19% Ge(Li) detector in anti-coincidence mode leads to the number of counts beneath the Compton continuum being reduced by a factor of 3.2. Suppression factors of 2 or 3 are typical in representative applications [93]. By splitting the signal from the 19% Ge(Li), and using two ADC's, two spectra may be simultaneously collected, one running in singles mode (ie unsuppressed) and the one running in anticoincidence mode (ie Compton suppressed). Gamma-rays which are totally absorbed in the Ge(Li) detector (and thus contribute to the FEP) are unaffected by the anticoincidence operation. Suppression of the Compton continuum in this way greatly facilitates measurement of the ^{210}Pb full energy peak.

3.4.4 Ge(Li) Efficiency

Gamma-ray photons resulting from the decay of radioactive nuclei within the sample will be emitted over the total 4π solid angle, ie in all directions from the sample. Thus, only a fraction of the γ-rays emitted from the sample will be incident upon the sensitive region of the detector. Of the γ-rays which are incident on the sensitive region only a fraction of these will undergo an interaction which results in a count in the full energy peak in the pulse-height spectrum. In order to obtain a value for the concentration of a radionuclide in a sample it is necessary to know the fraction of γ-rays emitted from the sample which will result in a count in the full energy peak. This fraction is known as the absolute efficiency of the full energy peak, $\varepsilon_{\text{full}}$.

The value of $\varepsilon_{\text{full}}$ varies with γ-ray energy. The maximum value is for γ-rays of energy $\sim 180\,\text{keV}$ and it falls away for γ-rays of higher and lower energy. The fall-off of $\varepsilon_{\text{full}}$ at higher energy is due to the fact that the cross-section for

a photoelectric interaction decreases as γ-ray energy increases [53], so that the probability of a γ-ray photon producing a count in the full energy peak is less for a higher energy γ-ray. For γ-rays of lower energy there is a greater possibility of absorption in the detector window or insensitive regions before reaching the sensitive region. The value $\varepsilon_{\text{full}}$ also depends critically on the sample-detector geometry and will be different for different detectors. By keeping the geometry constant and establishing the efficiency of each individual detector these problems may be overcome. In order to establish the efficiency curve for each detector a vegetation matrix mixed with a solution of radionuclides of known activity was used. The vegetation standard was prepared by ITE Merlewood using a mixed nuclide standard supplied by the National Physical Laboratory together with a ^{210}Pb solution supplied by LMRI, a part of CEA, the French Atomic Energy Commision. The standard produced a number of γ-rays of energy ranging from 46.5 keV to 1,800 keV and was presented to the Ge(Li) in exactly the same geometry as was used to analyse the samples.

The resulting efficiency curves for the 19% and 12% Ge(Li)'s are displayed in Figure 3.10. At higher energies the 19% Ge(Li) is more efficient than the 12% which is to be expected as it is has a larger active volume. However, at lower energies the 12% is more efficient. This is probably due to the 19% Ge(Li) having a slightly thicker window so a higher proportion of low energy γ-rays are absorbed before reaching the active volume and the efficiency at low energies is reduced overall. Detectors with larger active volumes are more efficient overall than detectors with small active volumes as they present a larger area and a greater thickness of the sensitive volume to the γ-rays from the sample. The way $\varepsilon_{\text{full}}$ varies with energy is dependent on the specific Ge(Li) and the sample-detector geometry used in the experiment. It is thus necessary to determine the full energy peak efficiency for each individual Ge(Li) detector, for the specific geometry to be used and for each γ-ray of interest, see Table 3.8.

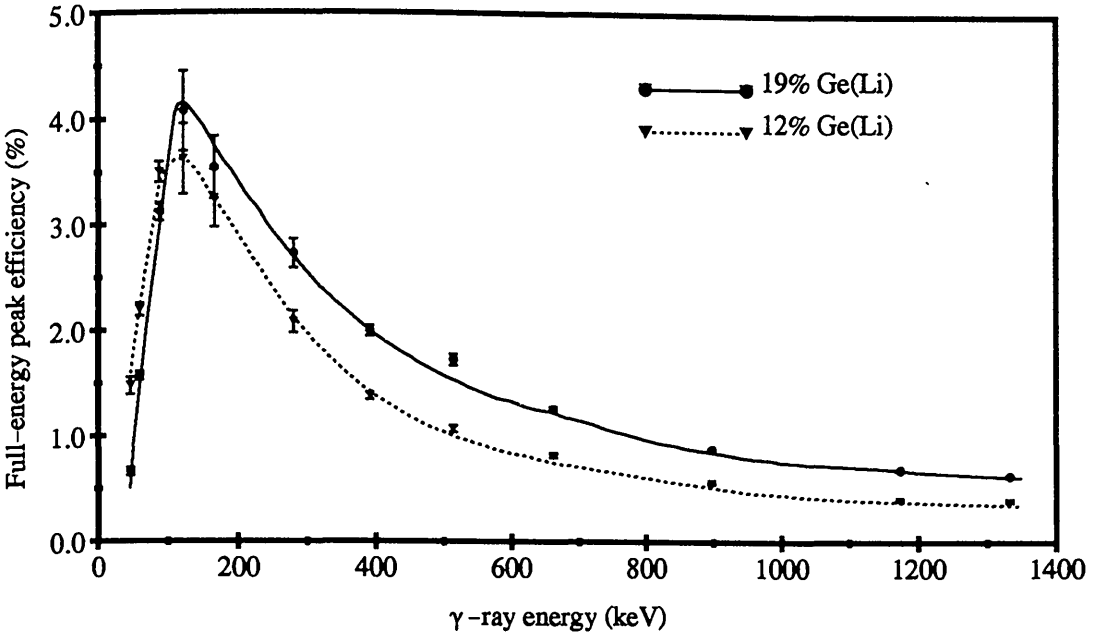

Figure 3.10: The full-energy peak efficiency curves for the two Ge(Li) detectors used in this study. The curves were obtained by counting a standard matrix which contained a variety of radionuclides, of known activity, emitting a range of γ-ray energies

3.4.5 Gamma-ray analysis of an unknown sample

Samples were analysed using two Ge(Li) detectors, a 19% model interfaced with a GEC 4190 computer using spectra analysis software developed at the Daresbury laboratory and a 12% model linked to a Canberra PC with an MCA simulator and using EG&G Ortec software.

The samples were presented to the detector in the top portion of a cylindrical perspex holder which fits neatly over the protective aluminium cap of the detector. Samples were counted usually for a period of 24 hours, though some samples were counted for longer periods to give improved statistics on the measured count rates. A typical γ-ray spectrum, obtained from a soil sample, is shown in Figure 3.11. At the end of the counting period, the following equation is used to give the activity of a specific nuclide in the counted sample:

$$A = \frac{N_p}{T} \times \frac{1}{\varepsilon_{full} \times b_\gamma} \times S$$

where A is the activity of the radionuclide in the counted sample in Becquerels (Bq), N_p is the number of counts in the full-energy peak, T is the period over which the counting took place in seconds, ε_{full} is the full-energy peak of the detector for the γ-ray of interest in the sample-detector geometry used and b_γ is the branching ratio of the radionuclide for the γ-ray used. The letter S represents the correction factor introduced to compensate for the self-absorption, within the sample, of low energy ^{210}Pb γ-rays.

Dividing the activity by the weight of the counted sample gives the specific activity ($Bq\,g^{-1}$). As the weight of material taken from the field and the surface area of the sample is known the results may be converted into $Bq\,cm^{-2}$ in the field. Thus, the inventory of the radionuclide of interest can be expressed as an activity per unit area. This is the best way of expressing the results, when considering deposition from the atmosphere, as it is independent of soil density, which varies between and within sites.

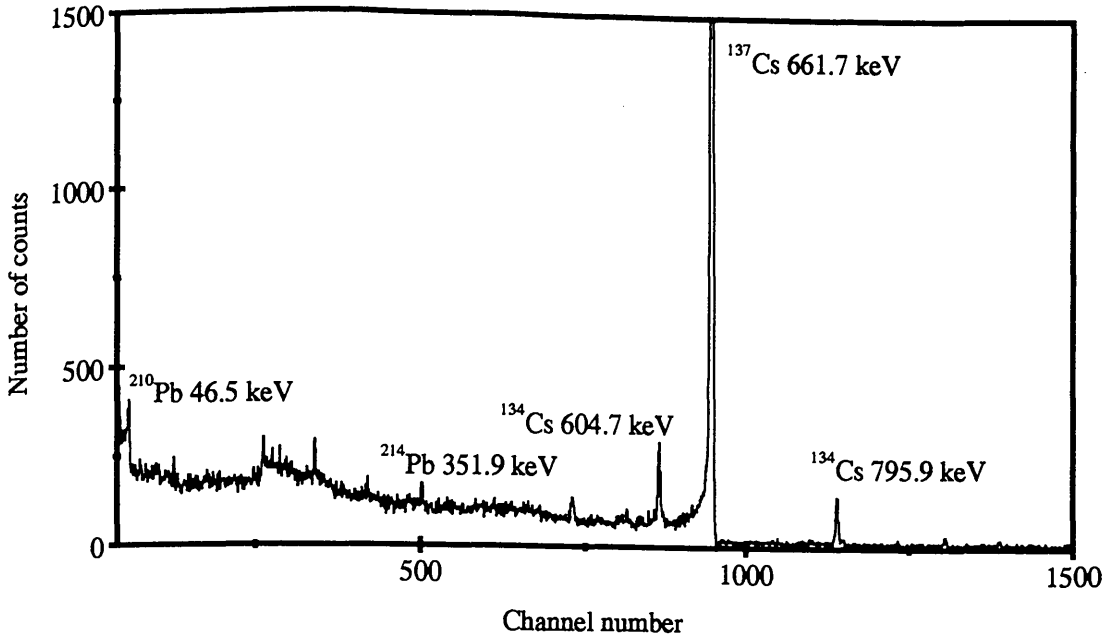

Figure 3.11: A γ-ray spectrum obtained with a soil sample and the 19% Ge(Li) Detector. The sample was collected from site L8 at Ben Lawers and represented the 1–5 cm depth interval

3.4.6 Radionuclides of interest

The radionuclides of interest in this study are listed in Table 3.8 along with the respective half-lives and branching ratios.

In measuring the activity of ^{210}Pb in the sample the low energy of the γ-ray produced in the decay of this nuclide meant that absorption within the sample itself could not be neglected. The weight of material in the standard holder varied between <20 g and >100 g depending on whether the sample was mainly vegetation or mainly mineral in character. Thus the density of the sample analysed varied by a factor of ~ 5. Following the technique of Cutshall et al. [40], samples were corrected for self-absorption, ie absorption of γ-rays within the sample itself, by placing a 0.03 μ Ci (1,110 Bq) ^{210}Pb source behind each sample and noting the proportion of the beam which penetrated the sample. This transmission (T) was then compared with the transmission through the Merlewood efficiency standard (I) to give a correction factor, S. Typically, the value of S lay between 1.0 and 1.5.

The radionuclide ^{226}Ra which is assumed to be in equilibrium with the 'sup-

Radionuclide	Half-life	γ-ray used (keV)	Branching ratio (%)
^{210}Pb	22.3 years	46.5	4.05
^{214}Pb	26.8 minutes	351.9	37.1
^{137}Cs	30.0 years	661.7	85.2
^{134}Cs	2.06 years	795.9	85.4
^{7}Be	53.3 days	477.6	10.4
^{228}Ac	6.13 hours	911.2	29.0
^{40}K	10^9 years	1460.8	10.7
^{208}Tl	3.05 minutes	2614.4	99.8

Table 3.8: Details of the radionuclides measured in the course of this study [16]

ported' ^{210}Pb produced within the soil, cannot be measured directly as the 186.1 keV γ-ray it produces occurs too closely to a ^{235}U γ-ray at 185.7 keV and the resolving power of the Ge(Li)'s was not sufficient to separate the contributions of these two γ-rays to the full energy peak. In order to estimate the 'supported' ^{210}Pb, equilibrium was assumed with ^{214}Pb which is present further down the U^{238} decay chain, see Figure 2.1, and produces a γ-ray at 351.9 keV.

The radionuclide ^{134}Cs was measured using the γ-ray at 795.9 keV. It was necessary to subtract the ^{228}Ac component from this peak as ^{228}Ac produces a γ-ray at 794.8 keV which cannot be separated from the ^{134}Cs peak. This ^{228}Ac contribution was estimated by measuring the peak at 911.2 keV and hence estimating the number of counts from this radionuclide which would occur in the ^{134}Cs 795.9 keV peak.

3.5 Uncertainties in the measured values

This section will attempt to quantify the uncertainties associated with the measured values, of the radionuclide inventories, presented in this study. Uncertainties

are introduced during, field sampling, sample preparation and during γ-ray analysis. In addition, at each sampling site there is an inherent variability of the radionuclide inventory. These four areas of uncertainty are investigated in turn.

Field sampling involved removing soil material from a fixed cross-sectional area. Maintaining areal control was not a problem when using a corer, but more difficult when excavating a pit. Great care was taken in excavating pits. Also, great care was taken to ensure that all the soil material, from the pit or core, was collected and assigned to the correct depth interval. It is difficult to quantify exactly the uncertainty introduced at this stage. However, a value of $\pm 10\%$ has been adopted to represent the uncertainty in the sampling procedure.

Preparation of the samples involved oven drying at $\sim 80°C$ and subsequent sieving or grinding. Again, great care was taken during preparation. Loss of soil material during sieving and grinding was small, and it is assumed that there was no preferential loss of any particular size fraction. Thus, uncertainties introduced at this stage are assumed to be insignificant in comparison to other sources of uncertainty.

At Ben Cruachan, Beinn Dorain and Ben Lawers 93, 95 and 78%, respectively, of the ^{210}Pb(atmos) inventory was measured in the top 5 cm of the soil profile. The measurements made on the 0–1 cm and 1–5 cm depth samples from Ben Cruachan will be used to illustrate the quantitative assessment of uncertainties involved in the γ-ray analysis. Sites C1 to C8 are represented, site C9 is excluded as it yielded an exceptional value for the ^{210}Pb(atmos) inventory. The uncertainty, ΔA, associated with the measured value of the activity, A, of a sample is a combination of the uncertainty in the number of counts in the full energy peak, N_p, the uncertainty in the full energy peak efficiency, ε_{full}, and the uncertainty in the correction factor introduced to compensate for self-absorption, S (in the case of ^{210}Pb only). The uncertainties associated with the period of counting, T, and the branching ratio, b_γ, are insignificant compared to the uncertainties in N_p, ε_{full} and S. For the 0–1 cm samples from Ben Cruachan the mean uncertainty in counting is $\pm 7\%$, for the 1–5 cm samples it is $\pm 17\%$. In both sets of samples the largest contributor

to the total counting uncertainty is the uncertainty in N_p, ie ΔN_p. The mean uncertainty of the 0–1 cm samples is less than for the 1–5 cm samples as the concentration of ^{210}Pb activity is greater in the surface samples. Thus, with a fixed counting period of 24 hours, the FEP for a surface sample will contain more counts than the FEP for the 1–5 cm sample ($\Delta N_p = N_p^{0.5}$). Combining the sampling and counting uncertainties gives mean uncertainties of $\pm 12\%$ and $\pm 20\%$ associated with the ^{210}Pb inventories of the 0–1 cm and 1–5 cm depth intervals, respectively, at Ben Cruachan. For the total ^{210}Pb(atmos) inventory at each site, C1–C8, at Ben Cruachan, the mean uncertainty is $\pm 10\%$. But how does this value for the average uncertainty associated with the total ^{210}Pb(atmos) inventory, due to the sampling and counting procedure, compare with the inherent variability of the ^{210}Pb(atmos) at a site ?

To investigate the intra-site variability, three or more points were sampled at certain sites. In addition, each sampling point represented either 2 or 5 individual cores, depending on whether the large or small corer was used, further taking into account intra-site variations. The variability may be quantified using the coefficient of variation (cov). At Great Dun Fell the mean cov was 29%, at Beinn Dorain it was 14% and at Ben Lawers it was 18%. Thus, the intra-site variability is greater than the uncertainty in the ^{210}Pb(atmos) inventory due to sampling and analysis, for the majority of sites in this study. It is probably wise to adopt an uncertainty of $\pm 20\%$ on the ^{210}Pb(atmos) inventory from any site at which only a single point was sampled.

Turning to the ^{137}Cs(bomb) results, the mean uncertainty associated with the ^{137}Cs(bomb) inventory at Ben Cruachan, due to sampling and analysis, is $\pm 8\%$. The cov was 19% at Great Dun Fell, 33% at Beinn Dorain and 14% at Ben Lawers. The mean cov is large for Beinn Dorain as at two of the sites there were points which gave outstandingly high values of the ^{137}Cs(bomb) inventory. Again, the intra-site variability is larger than the uncertainty due to sampling and analysis, and an uncertainty of $\pm 20\%$ on the value of ^{137}Cs(bomb) from any site at which only one point was sampled seems prudent.

3.6 Loss-on-ignition Experiments

All the samples collected from the Merrick and the samples collected from sites C5 and C9 at Ben Cruachan were subjected to loss-on-ignition (LOI) experiments. This involved weighing the sample before placing it in a furnace for about 12 hours at $\sim 500\,^{\circ}\mathrm{C}$. Any organic matter in the sample will ignite. Re-weighing the sample after removal from the furnace enables the weight loss in the furnace to be calculated. The weight loss is taken to correspond to the organic matter content of the sample. The organic component is usually expressed as a percentage of the weight of the original soil sample. However, the loss on ignition will also include a contribution from clay dehydration. In low-clay high-organic soils the contribution due to clay dehydration will probably be insignificant. In the more mineral soils the values for the organic matter component may be overestimates. This is not too serious as an indication, within a few %, of the organic matter content is desired for the purposes of this study rather than a precise value. Nozaki et al. found a strong linear correlation, within each profile, between 'excess' ^{210}Pb per gram of dry soil and the mass loss on ignition [117]. They also found the relationship to hold regionally, for Pennsylvania soils.

3.7 Rainfall Data

The annual rainfall at each of the 66 individual sampling sites was estimated from the Meteorological Office Map of Average Annual Rainfall, for the international standard period 1941–1970 [107]. The map represents the annual rainfall as a contour map and was compiled using measured data from rain gauges, and then contouring and extrapolating rainfall through height-rainfall relationships. It is thought that the map compilers based their map on the underlying topography to extrapolate from raingauges up to higher altitudes and applied a rainguage reading to the same height in the same area. There is, therefore, a heavy dependence of the map on the underlying topography.

To obtain a value for the average annual rainfall at each of the 66 sites their position was pinpointed on the average annual rainfall map and a value estimated by interpolating between the rainfall contours. This only gives a rough estimate of the rainfall at each site, but it is probably accurate to within $\sim 10\%$ at the lower altitude sites and $\sim 30\%$ at higher altitudes.

Figure 1.3 shows the average annual rainfall for Britain and Ireland. The high rainfall amounts falling in the upland regions of northern and western Britain compared to the east and south can clearly be seen.

Chapter 4

Presentation of Results

In this chapter, the radioanalytical and other results for each mountain location are presented. Appropriate sub-sets of the data are discussed in detail in later chapters. Specifically, the results presented in this chapter are:

1. inventories of each radionuclide of interest at each site,
2. data for the distribution of ^{210}Pb(atmos), ^{137}Cs(bomb) and ^{134}Cs down soil profiles on the Merrick;
3. loss-on-ignition results for all the Merrick samples and selected ones from Ben Cruachan; and
4. variation of rainfall with altitude at each mountain.

4.1 Radionuclide Inventories

For each mountain, the inventories of atmospherically derived ^{210}Pb, termed ^{210}Pb(atmos), total ^{137}Cs, termed ^{137}Cs(tot), ^{137}Cs in nuclear weapons fallout, termed ^{137}Cs(bomb), ^{137}Cs due to Chernobyl deposition, termed ^{137}Cs(Chern) and ^{134}Cs are presented. In addition results for ^{7}Be are given for the sites sampled at the Merrick on 26^{th} Oct 1988 (sites M1–M6).

The inventory of each radionuclide is given in units of $\mathrm{Bq\,cm^{-2}}$ so that the data are independent of soil density. The conversion of the results from units of

$Bq\,kg^{-1}$, obtained by dividing the activity of the sample by its mass, to $Bq\,cm^{-2}$ is achieved by recording the mass of material removed from each depth interval in the field and the cross sectional area of the field sample (ie the core or pit area). The values in Bq cm^{-2} for each depth interval are summed to give the total inventory to a defined depth in the field.

The ^{210}Pb(atmos) values were obtained by subtracting the ^{214}Pb activity in the sample from the total ^{210}Pb activity. This procedure assumes equilibrium between the activity of ^{214}Pb and the activity of the 'supported' ^{210}Pb in the sample and is reasonable since the two intermediate isotopes, ^{214}Bi and ^{214}Po, are also short-lived. The total ^{137}Cs in each sample is separated into two fractions, that due to the weapons testing and that due to the Chernobyl accident. The quantity of ^{137}Cs(Chern) in each sample is obtained by multiplying the ^{134}Cs activity[1] by the factor of 1.82 which is the ratio of ^{137}Cs : ^{134}Cs measured in rainfall at Eskdalemuir after the radioactive plume reached Britain [23]. The ^{137}Cs(bomb) fraction in each sample is obtained by subtracting the ^{137}Cs(Chern) from the total ^{137}Cs present.

4.1.1 Merrick

The inventories of ^{210}Pb(atmos), ^{137}Cs(bomb), ^{134}Cs, ^{137}Cs(tot), ^{137}Cs(Chern) and ^{7}Be are presented in Figures 4.1 to 4.4. Figures 4.1 and 4.2 show the results from pit sampling, one pit being excavated at each of the 6 sites sampled on the 26^{th} Oct 1988. As only one pit was taken the error bars associated with the points represent a combination of the uncertainty involved in determining the count-rate of each radionuclide (governed by Poisson Statistics) and the uncertainty in determining the efficency of detection, ϵ_{full}. The dotted line represents the hill profile as it does in the plots of the inventories for each of the 5 mountains.

Figures 4.3 and 4.4 show the results for the same set of radionuclides from the core samples taken at each of the original 6 sites and also from the cores taken

[1] decay corrected back to the reference date of 2^{nd} May 1986

from sites M7 and M8, sampled on 30^{th} Nov 1990 and represented by black circles on the figures. The inventories of ^{7}Be are not shown for sites M7 and M8 as the long separation between sampling dates and the short (53.3 day) half-life of ^{7}Be invalidates plotting the results on the same graph. Three sets of cores were taken from sites M1–M6 and two sets of cores from sites M7 and M8. The error bars on Figures 4.3 and 4.4 represent either the fractional error (ie the uncertainty involved when combining several values, each of which has a measurable uncertainty) or the standard error on the mean, whichever proved to be the largest. Wherever several values have been combined this approach has also been adopted.

In the case of ^{210}Pb(atmos) the measured inventories from the pit and core samples taken from sites M1–M6 show an increase with altitude. There appears to be a factor increase of 2–3 between the values from sites M6 and M5 and the values from sites M4 to M1. At low levels the inventory has a mean value of about $0.2\,\mathrm{Bq\,cm^{-2}}$ and approaches $0.5\,\mathrm{Bq\,cm^{-2}}$ at the higher sites. The inventory measured from the pit sample at M3 is lower than would be expected. However, sites M7 and M8 which were sampled more than 2 years after the original expedition gave unexpected results. These sites although lying between M6 and M5 show measured inventories of ^{210}Pb(atmos) closer to those measured at the higher altitude sites and this result will be discussed in detail in section 5.1.1.

The inventories of ^{137}Cs(bomb) show an increase with altitude and the values measured from the pit and core samples are a good deal smaller for M6 than for M5 to M6. The value from the pit sample at M3 is again lower than expected as is the pit sample from M6. The core samples from M5 and M6 reveal a possible decrease of the ^{137}Cs(bomb) inventory at higher altitudes. The new sites M7 and M8 give results more in line with the old sites but are again slightly higher than the neighbouring sites, M5 and M6 respectively. As the ^{137}Cs(bomb) values are obtained by subtracting the Chernobyl component from the total ^{137}Cs present, and at the Merrick the Chernobyl component is a large fraction of the total ^{137}Cs present, the error bars are relatively large. The Chernobyl deposition of ^{137}Cs at the Merrick amounted to roughly three times the amount already present due to

nuclear weapons testing.

The inventories of ^{134}Cs from the pit samples show a hint of an increase with altitude and again the pit at M3 shows a lower value than expected. Looking at the values from the cores no increase with altitude is indicated. The diagrams of the ^{137}Cs(tot) and ^{137}Cs(Chern) inventories contain no extra information on the pattern of ^{137}Cs distribution over the hill but are useful to show the values at the site. The results from the pits and the cores both show an increase in the ^{7}Be inventory with altitude. A greater rate of increase is indicated from the pits than from the cores.

The distributions of ^{210}Pb(atmos), ^{137}Cs(bomb) and ^{134}Cs down the sampled soil profiles are given in Figures 4.5 to 4.13. The depth profiles for each radionuclide are given first for the pit samples then for the core samples and lastly for the two sites sampled later, M7 and M8. Figures 4.5 to 4.7 show the depth profiles for ^{210}Pb(atmos), the depth profiles for ^{137}Cs(bomb) are shown in Figures 4.8 to 4.10 and those for ^{134}Cs are shown in Figures 4.11 to 4.13. The results are presented as the distribution of activity per unit volume (Bq cm^{-3}) with depth (cm) down the profile.

At each of the 8 sites sampled no significant amounts of ^{210}Pb(atmos) were detected below 10 cm depth. Considering sites M1–M6 the greater part of the ^{210}Pb(atmos) inventory was found in the 0–5 cm sections of the pit samples. The depth profiles from the core samples show the much greater concentration of ^{210}Pb(atmos) in the 0–1 cm depth interval than in the 1–10 cm depth interval for all the sites except site M6. The results from the pit and core samples at site M6 indicate that the highest concentration of ^{210}Pb(atmos) is present between 1–5 cm depth. Most of the ^{210}Pb(atmos) at sites M7 and M8 is present in the top 5 cm of the soil profile.

The depth profiles of ^{137}Cs(bomb) for sites M1–M6 indicate deeper penetration than for ^{210}Pb(atmos). A greater proportion of ^{137}Cs(bomb) is found in the 5–10 cm and 1–10 cm depth intervals than is the case for ^{210}Pb(atmos). However, the evidence from the 6 pit samples shows that most of the ^{137}Cs(bomb) detected

down to 20 cm depth is present in the 0–10 cm section. The depth profiles of ^{137}Cs(bomb) for sites M7 and M8 are interesting to compare. Whereas most of the ^{137}Cs(bomb) at site M7 is present in the upper 5 cm and the concentration falls off sharply with depth, at site M8 the ^{137}Cs(bomb) is spread much more evenly down the profile. The depth profiles for sites M1–M8 show that ^{134}Cs is present within a few cm of the surface and that most lies above 5 cm depth.

4.1.2 Great Dun Fell

The radionuclide inventories of ^{210}Pb(atmos), the various ^{137}Cs fractions and ^{134}Cs from Great Dun Fell are given in Figs 4.14 and 4.15. Each cross in Figures 4.14 and 4.15 represents the combination of measurements from 3 sampling points with the exception of sites G3 and G7, at which samples were collected from one point, and site G20 at which samples were collected from two points. Site G8 was underneath a small stand of conifers and is given the symbol of a black square to highlight its different status.

A clear increase is seen in the inventory of ^{210}Pb(atmos) at Great Dun Fell between the Eden Valley and the summit. The mean of the lowest 6 sites, excluding site G8, is $< 0.2\,\mathrm{Bq\,cm^{-2}}$ and at the summit values approach $0.6\,\mathrm{Bq\,cm^{-2}}$, a factor increase of ~ 3. To the east downwind of the summit the measured inventories continue to be more than a factor of 2 greater than the mean of those in the Eden valley.

The pattern of ^{137}Cs(bomb) is more complex. There is an apparent increase with altitude between the valley and the summit of about a factor of 2 and the highest values of the inventory are observed at the summit. Some sites, however, particularly to the east of the summit show very low inventories. The largest ^{134}Cs inventories are observed at the summit and the smallest in the Eden Valley. The deposition of ^{137}Cs at Great Dun Fell from the Chernobyl accident amounted to about 10% of that already present due to nuclear weapons testing.

4.1.3 Ben Cruachan

The radionuclide inventories for Ben Cruachan are plotted in Figures 4.16 and 4.17. At each of the 9 sites sampled on this mountain, except at site C5, a single point was sampled. The error bars represent the uncertainty associated with the counting statistics and the uncertainty associated with $\varepsilon_{\text{full}}$. At site C5, samples were collected from two points. As the measured inventories of ^{210}Pb(atmos) and ^{137}Cs(bomb) at these two sampling points were so different they were not combined and each is represented separately. The measurements at this site will be discussed in section 5.1.3.

The measurements of the ^{210}Pb(atmos) inventory at Ben Cruachan show an increase of a factor of just under 3 between sites C7 and C8 and site C2. Site C9 gave unfeasibly small values for the ^{210}Pb(atmos) and ^{137}Cs(bomb) inventories and no values were obtained lower than 350 m asl, ie the altitude of site C8. Thus it is possible that the factor increase in the ^{210}Pb(atmos) inventory is greater than shown. Site C2, downwind of the summit, shows the largest ^{210}Pb(atmos) inventory at 1.05 Bq cm^{-2}. This is the largest value of all 66 sites sampled in this study.

The inventory of ^{137}Cs(bomb) at Ben Cruachan shows an increase of about a factor of 3 between the lower altitude sites and the higher altitude sites. No clear pattern is apparent in the measurements of the ^{134}Cs inventory over Ben Cruachan. Deposition of ^{137}Cs(Chern) at Ben Cruachan was relatively slight, accounting for about 10% of the ^{137}Cs already present due to weapons testing.

4.1.4 Beinn Dorain

The radionuclide inventories obtained from the 11 sites sampled at Beinn Dorain are presented in Figures 4.18 and 4.19. Three points were sampled at sites D1, 4, 7 and 11 and at each of the other sites a single point was sampled. Two of the sites sampled D5 and D6 were on a very steep incline.

The inventory of ^{210}Pb(atmos) over Beinn Dorain shows an increase of a factor

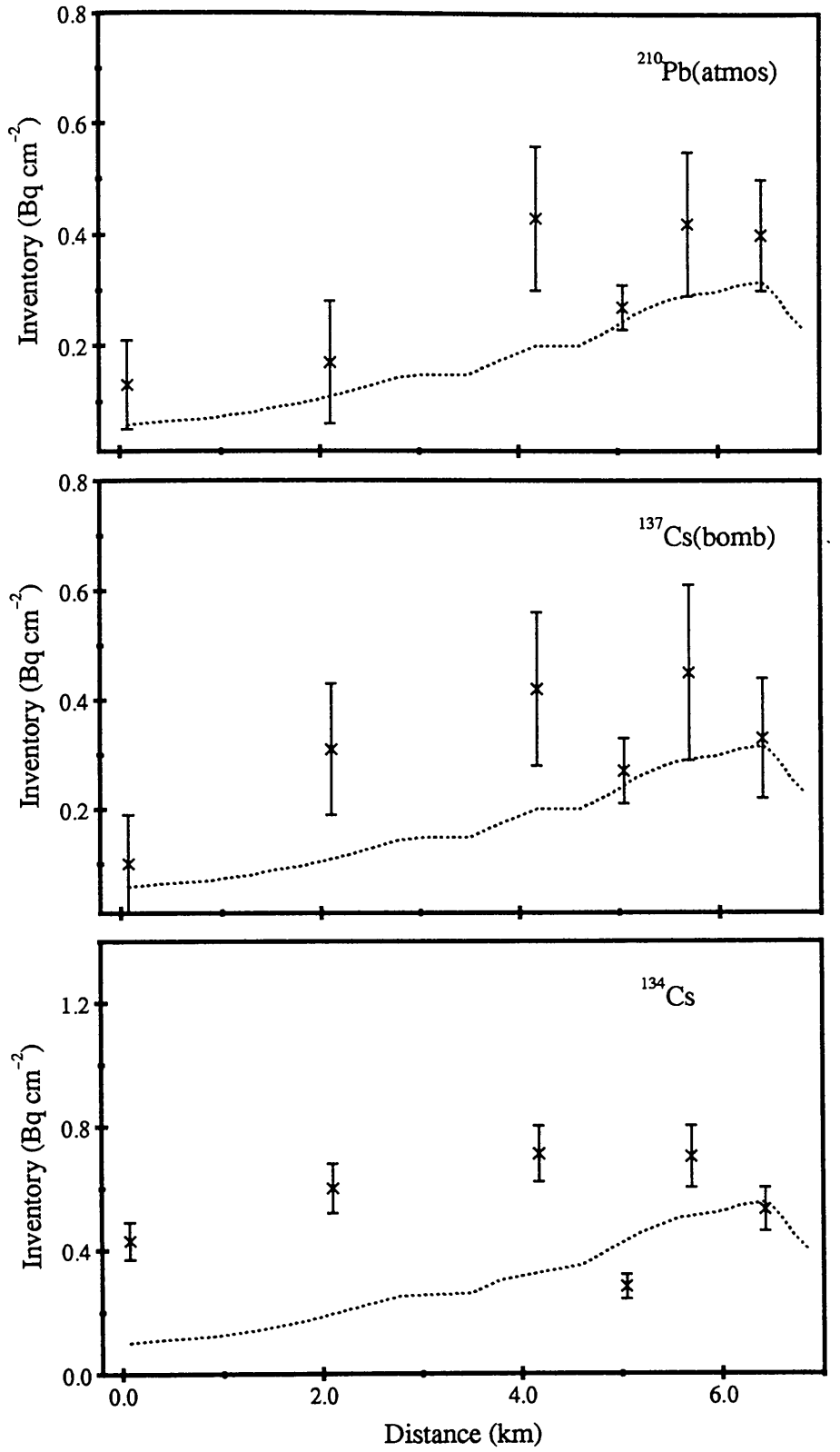

Figure 4.1: ^{210}Pb(atmos), ^{137}Cs(bomb) and ^{134}Cs inventories measured in the Pit samples at the Merrick

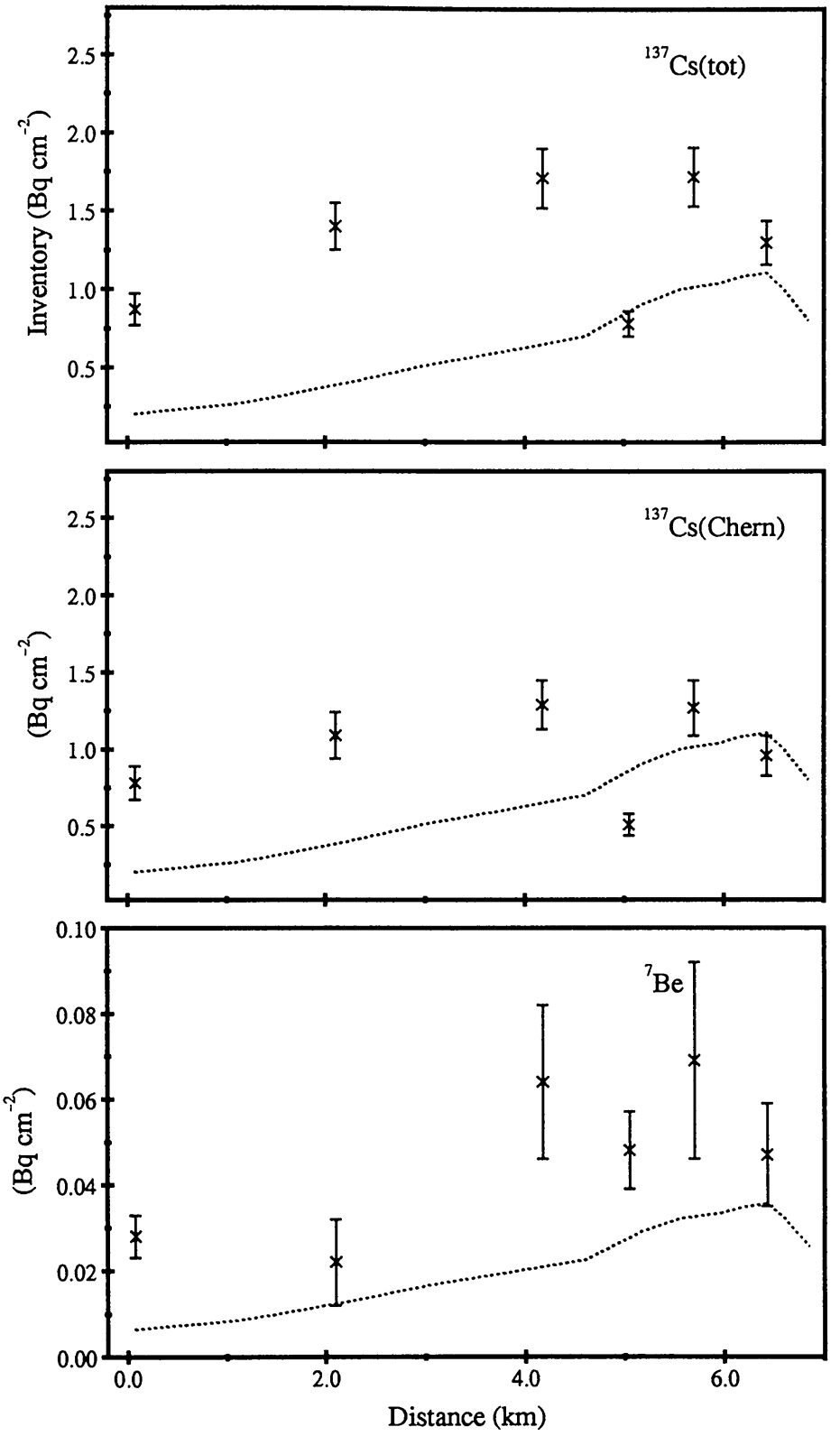

Figure 4.2: ^{137}Cs(tot), ^{137}Cs(Chern) and ^{7}Be inventories measured in the Pit samples at the Merrick

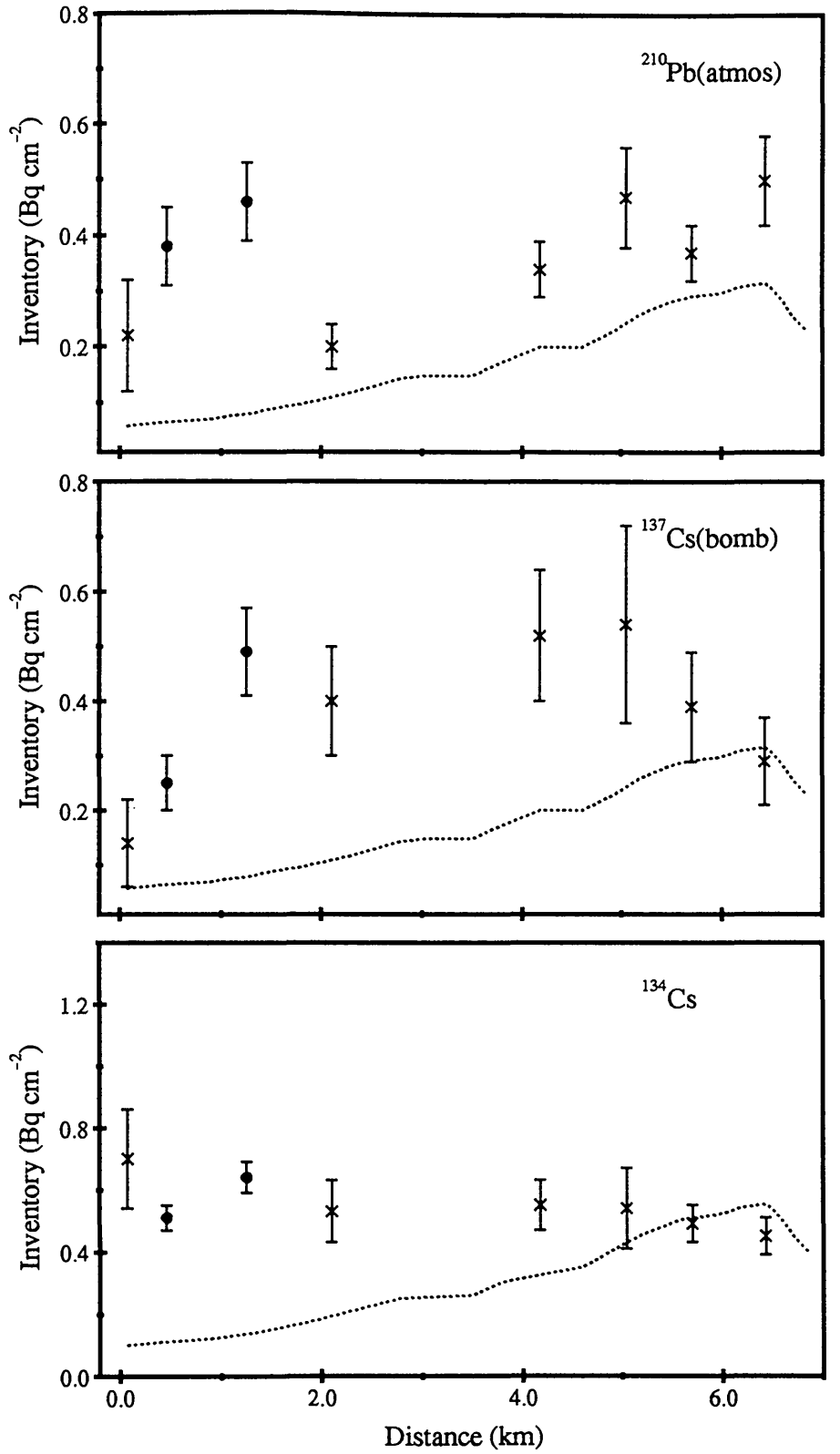

Figure 4.3: ^{210}Pb(atmos), ^{137}Cs(bomb) and ^{134}Cs inventories measured in the Core samples at the Merrick

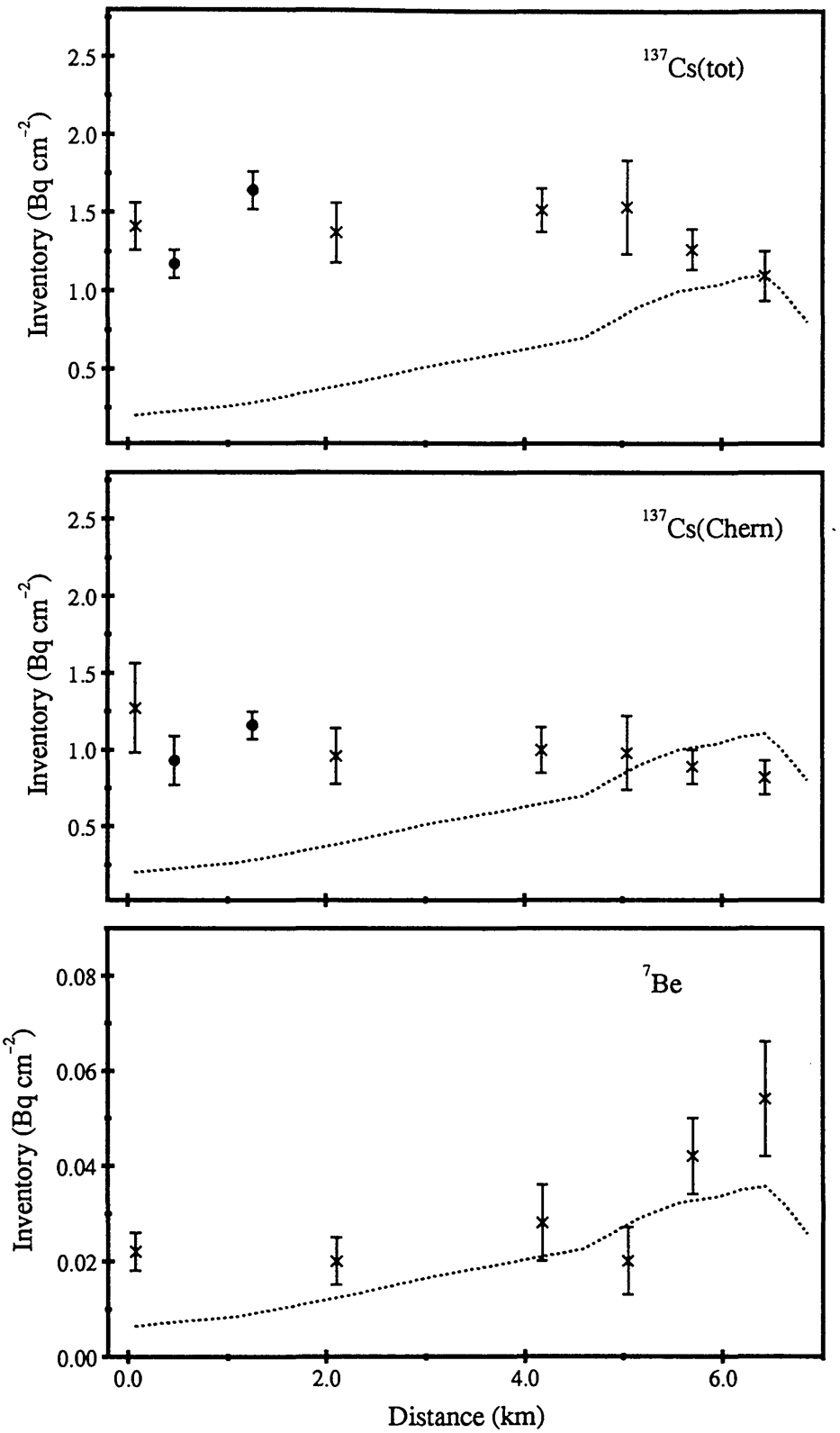

Figure 4.4: ^{137}Cs(tot), ^{137}Cs(Chern) and ^{7}Be inventories measured in the Core samples at the Merrick

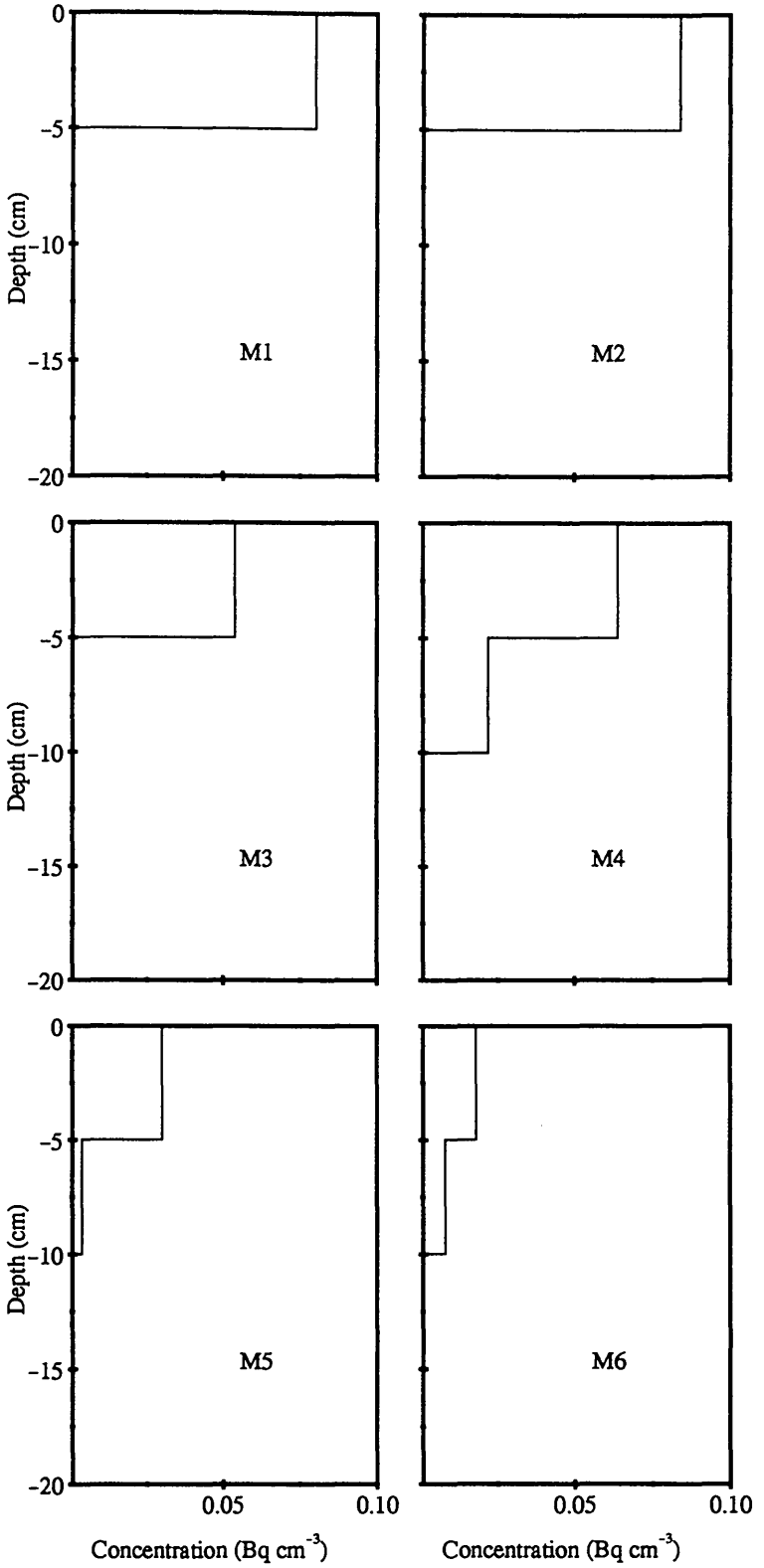

Figure 4.5: Distribution of ^{210}Pb(atmos) with depth at the Merrick. Pit Samples, sites M1–M6

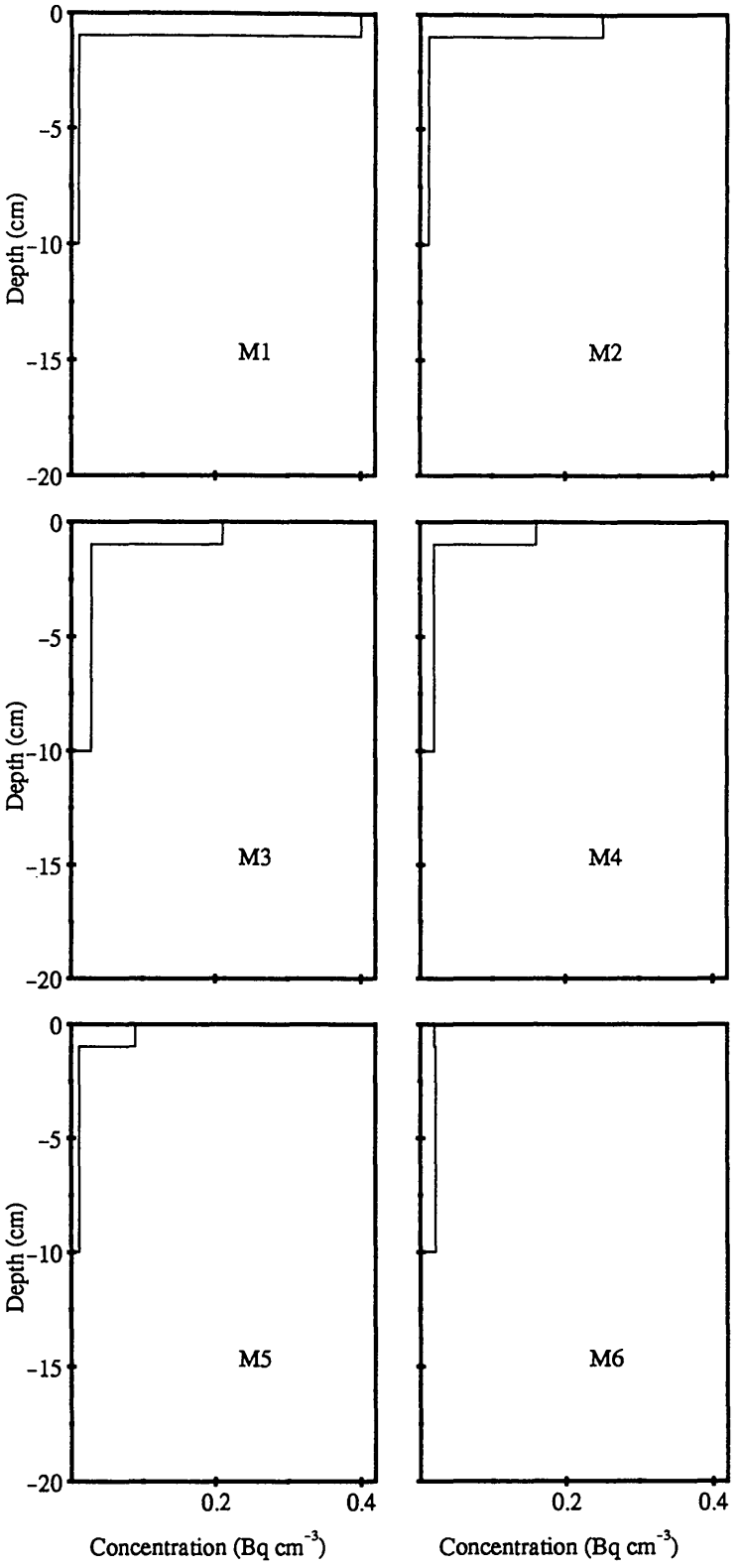

Figure 4.6: Distribution of ^{210}Pb(atmos) with depth at the Merrick. Core Samples, sites M1–M6

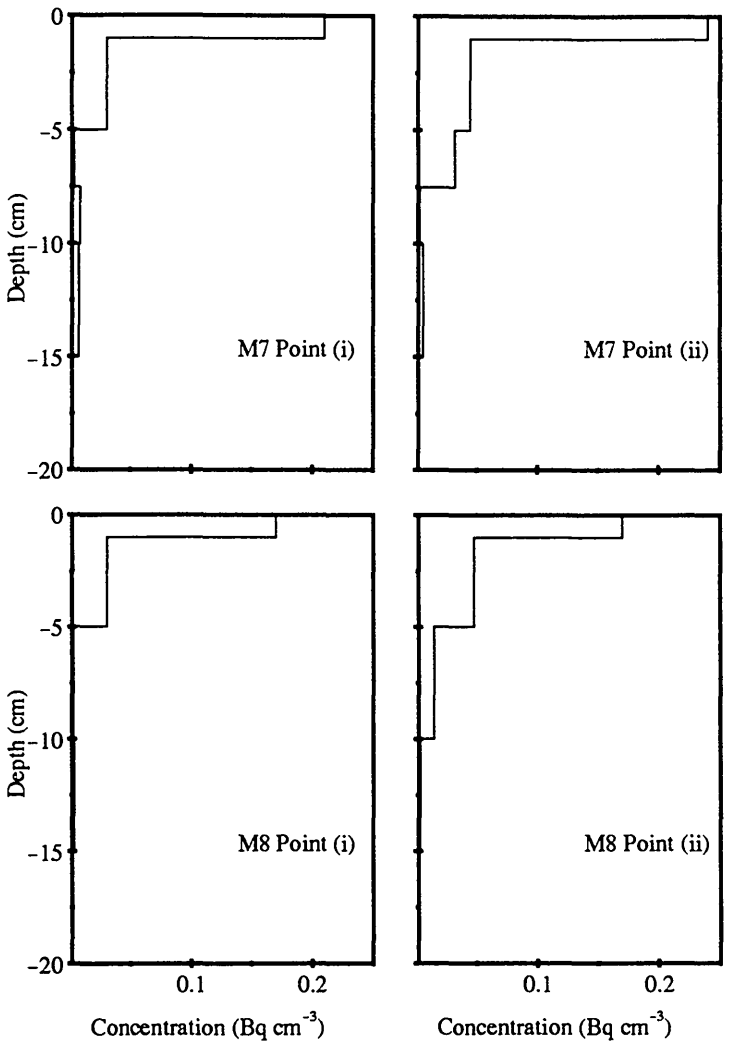

Figure 4.7: Distribution of ^{210}Pb(atmos) with depth at the Merrick. Sites M7 and M8

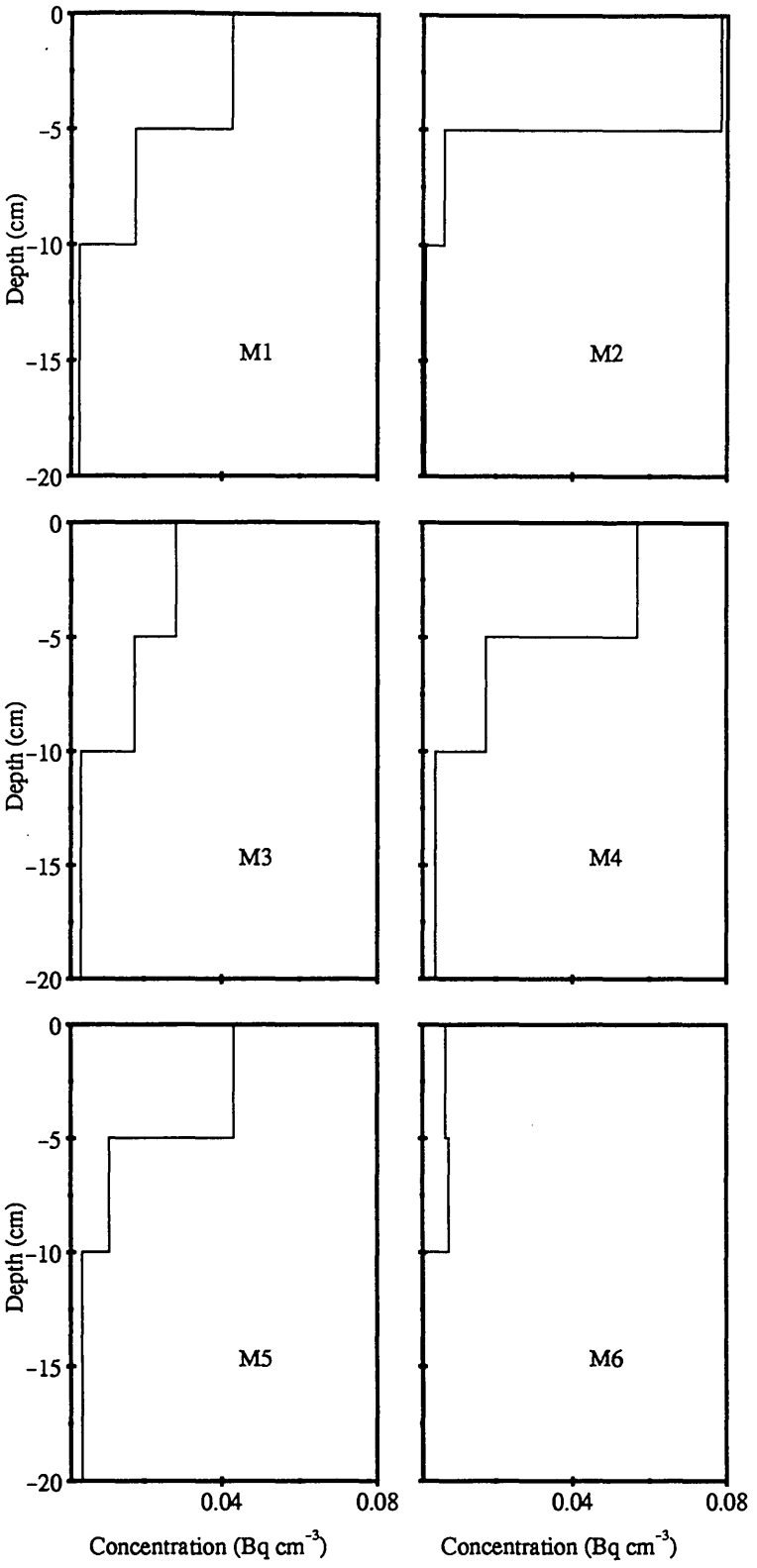

Figure 4.8: Distribution of ^{137}Cs(bomb) with depth at the Merrick. Pit Samples, sites M1–M6

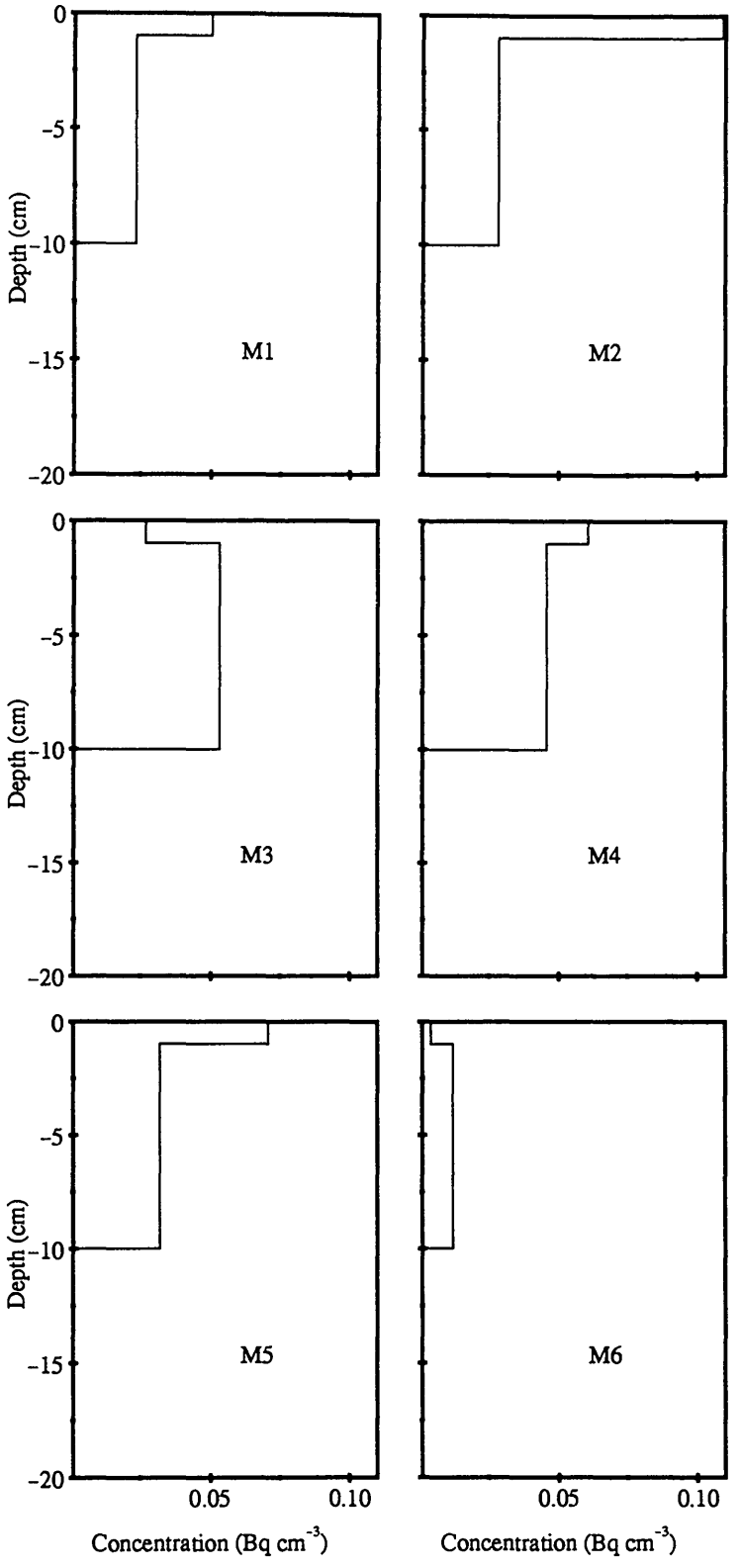

Figure 4.9: Distribution of ^{137}Cs(bomb) with depth at the Merrick. Core Samples, sites M1–M6

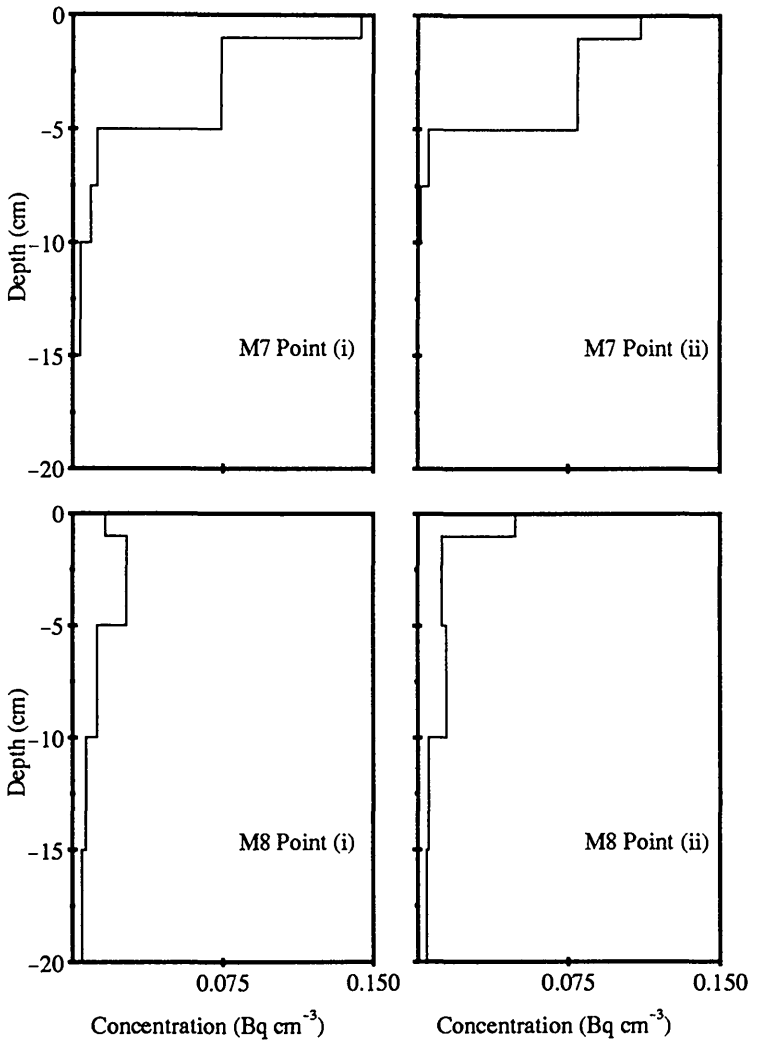

Figure 4.10: Distribution of ^{137}Cs(bomb) with depth at the Merrick. Sites M7 and M8

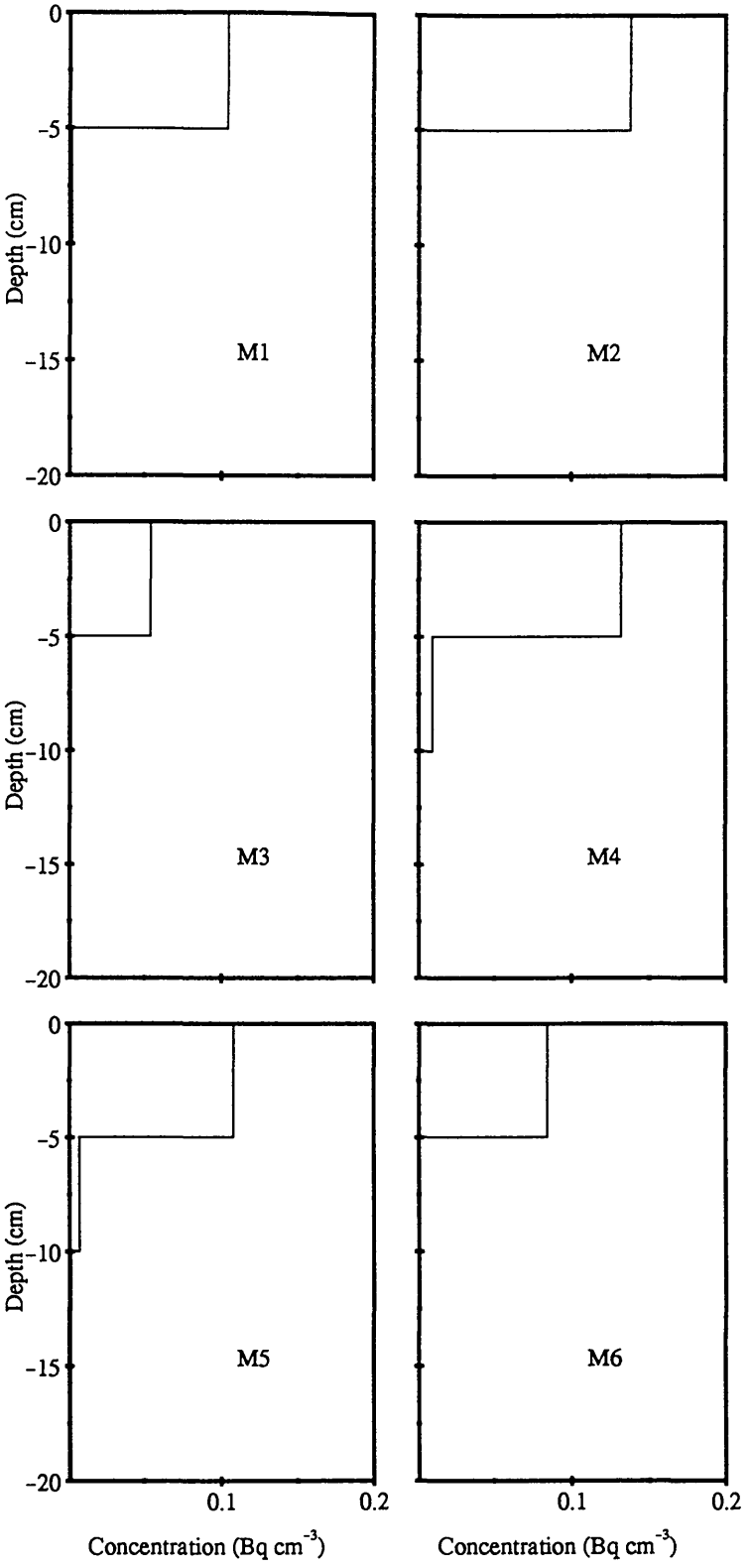

Figure 4.11: Distribution of ^{134}Cs with depth at the Merrick. Pit samples, sites M1–M6

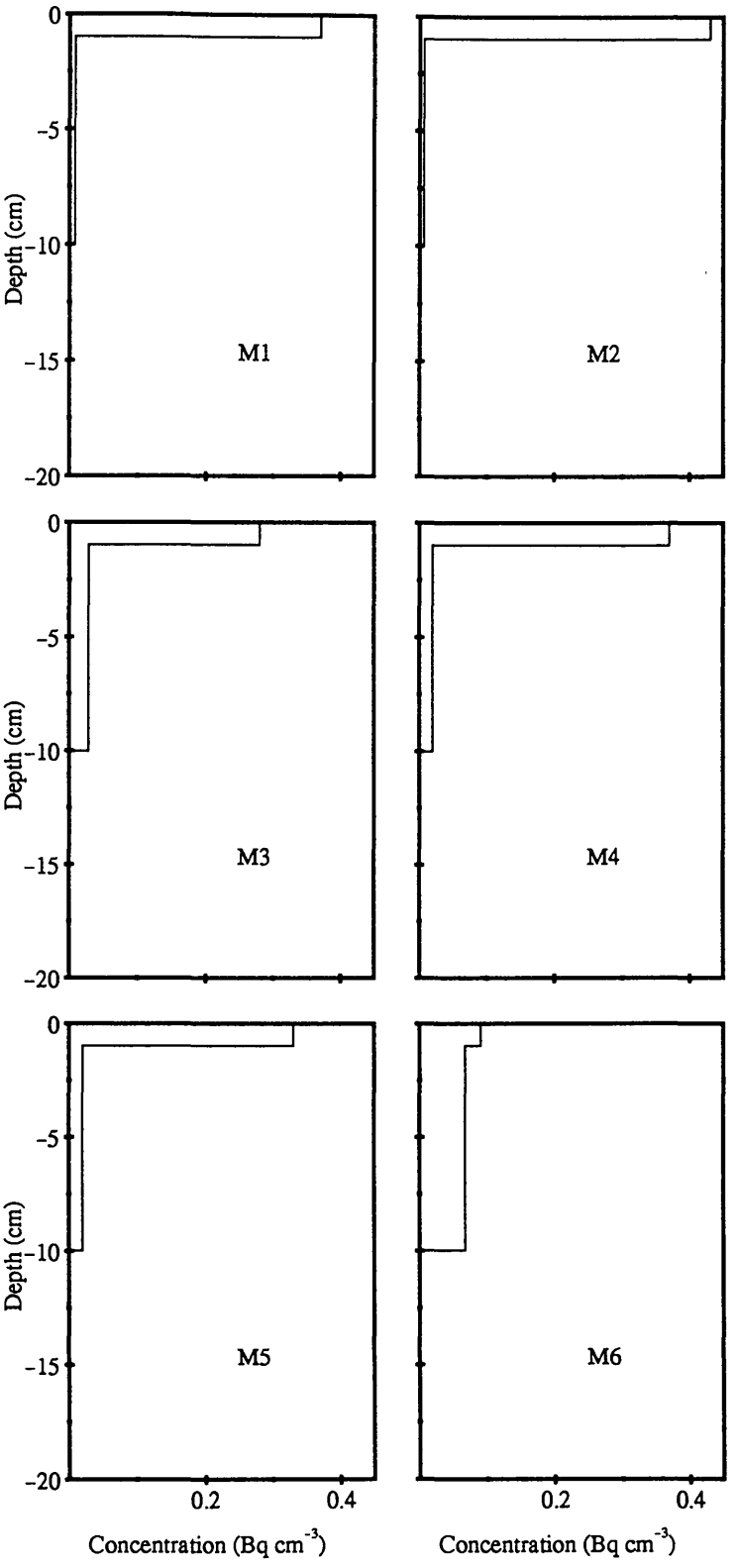

Figure 4.12: Distribution of ^{134}Cs with depth at the Merrick. Core Samples, sites M1–M6

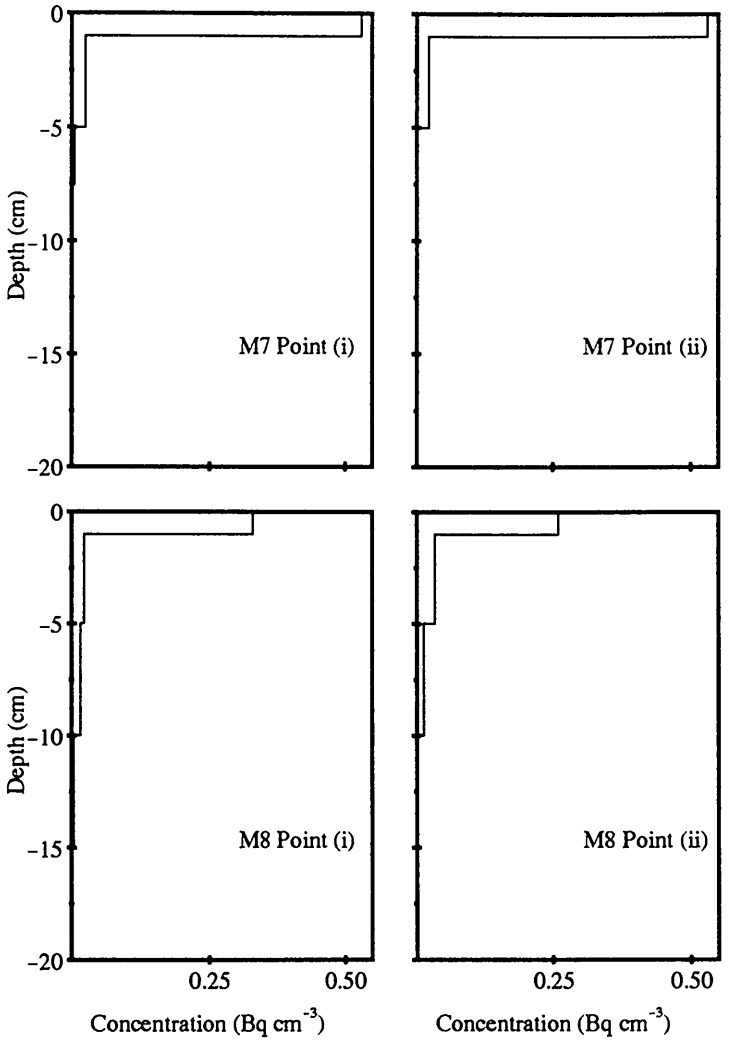

Figure 4.13: Distribution of ^{134}Cs with depth at the Merrick. Sites M7 and M8

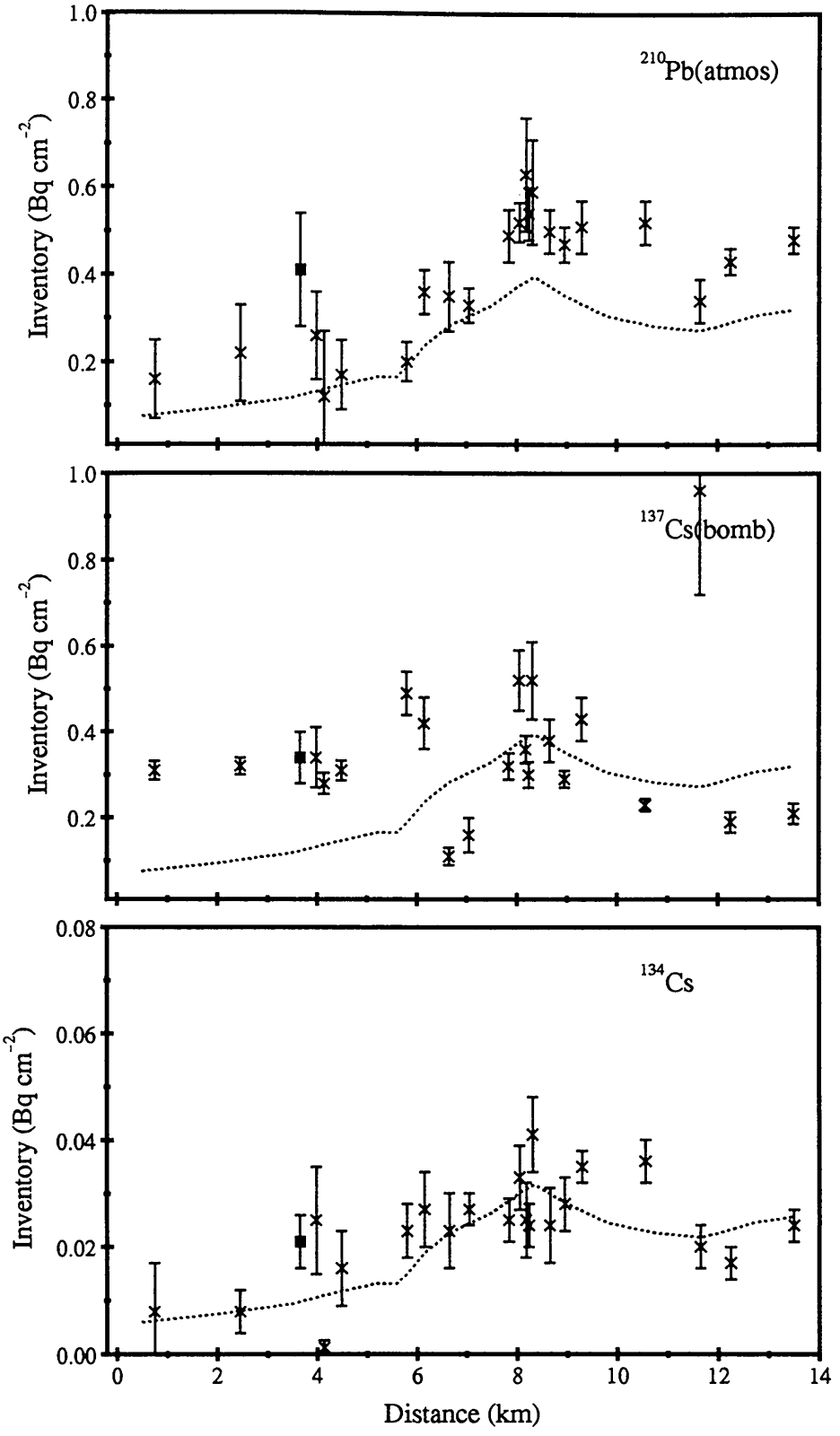

Figure 4.14: ^{210}Pb(atmos), ^{137}Cs(bomb) and ^{134}Cs inventories measured at Great Dun Fell

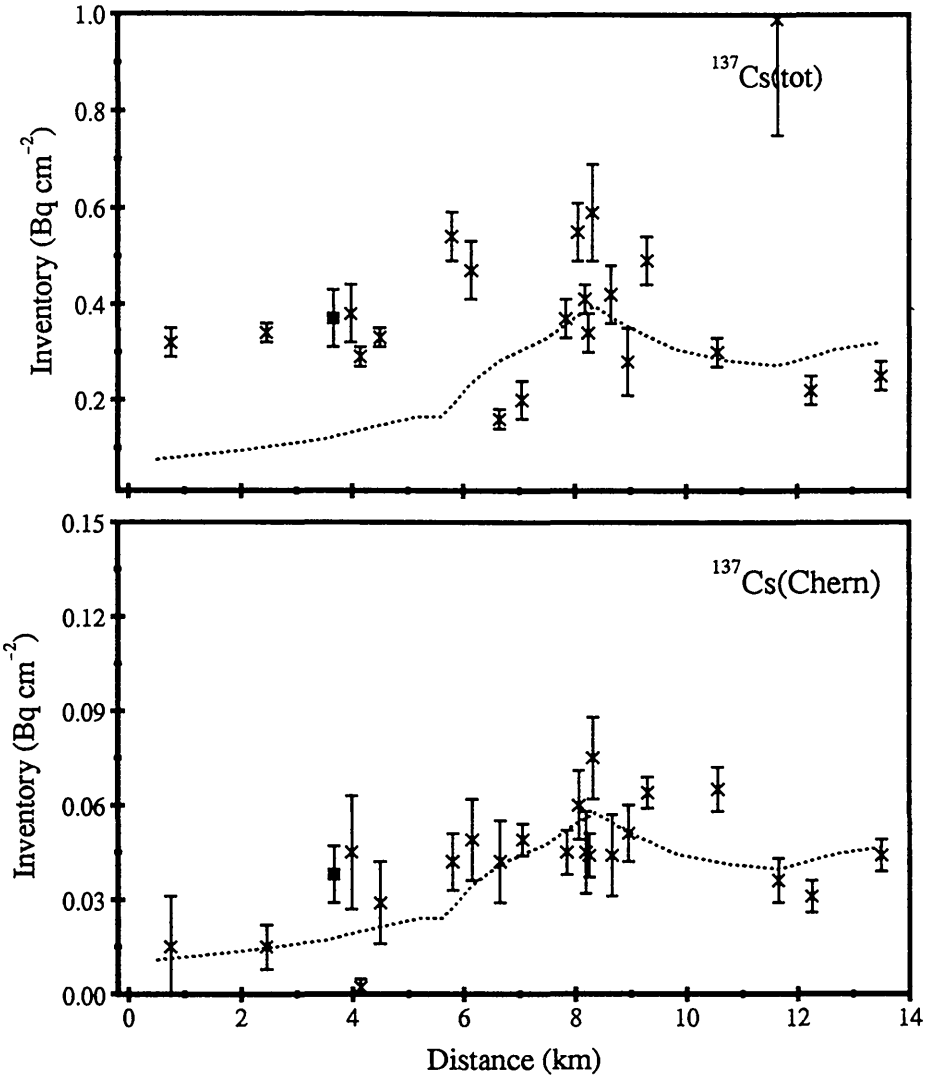

Figure 4.15: ^{137}Cs(tot) and ^{137}Cs(Chern)inventories measured at Great Dun Fell

of ~ 2 between the lowest sites in the valley and the highest sites. The increase does not appear to be a smooth one though. An increase with altitude is also seen in the ^{137}Cs(bomb) inventory. Sites D2, 3 and 4 seem a little lower than would be expected. The inventory of ^{134}Cs shows a clear increase with altitude of a factor of ~ 4 between the valley and site D2. Site D1 at the summit has a very low value. The rate of increase appears to follow the hill profile very closely. The mean inventory of ^{137}Cs(Chern) represents 46% of that of ^{137}Cs(bomb) present.

4.1.5 Ben Lawers

Figures 4.20 and 4.21 show the radionuclide inventories for the 15 sites sampled at Ben Lawers. Two points were sampled at site L8 and three at sites L10, 13, 14 and 15. Four points were sampled at site L12. At each of the other sites a single point was sampled.

Of all the 5 mountains sampled in this study Ben Lawers shows the smallest increase in the ^{210}Pb(atmos) inventory with altitude despite the substantial height range over which sampling took place. The lowest lying 6 sites (excluding site L11) give a very consistent value of the ^{210}Pb(atmos) inventory of $\sim 0.3\,\mathrm{Bq\,cm^{-2}}$ but at higher altitudes the inter-site variability increases.

The measured ^{137}Cs(bomb) inventories appear to be highly variable with no clear pattern. Site to site variability is high over most of the height range. Interestingly sites L15 to L12 show a possible reduction in the measured ^{137}Cs(bomb) inventories with altitude. The error bars are large at some of the sites due to the fact that deposition of ^{137}Cs(Chern) was substantial over Ben Lawers amounting to 4–5 times the ^{137}Cs(bomb) already present. The measured values of the ^{134}Cs inventory are very consistent at $\sim 0.9\,\mathrm{Bq\,cm^{-2}}$ for sites L15 to L6 but at higher altitudes the site to site variation is very large.

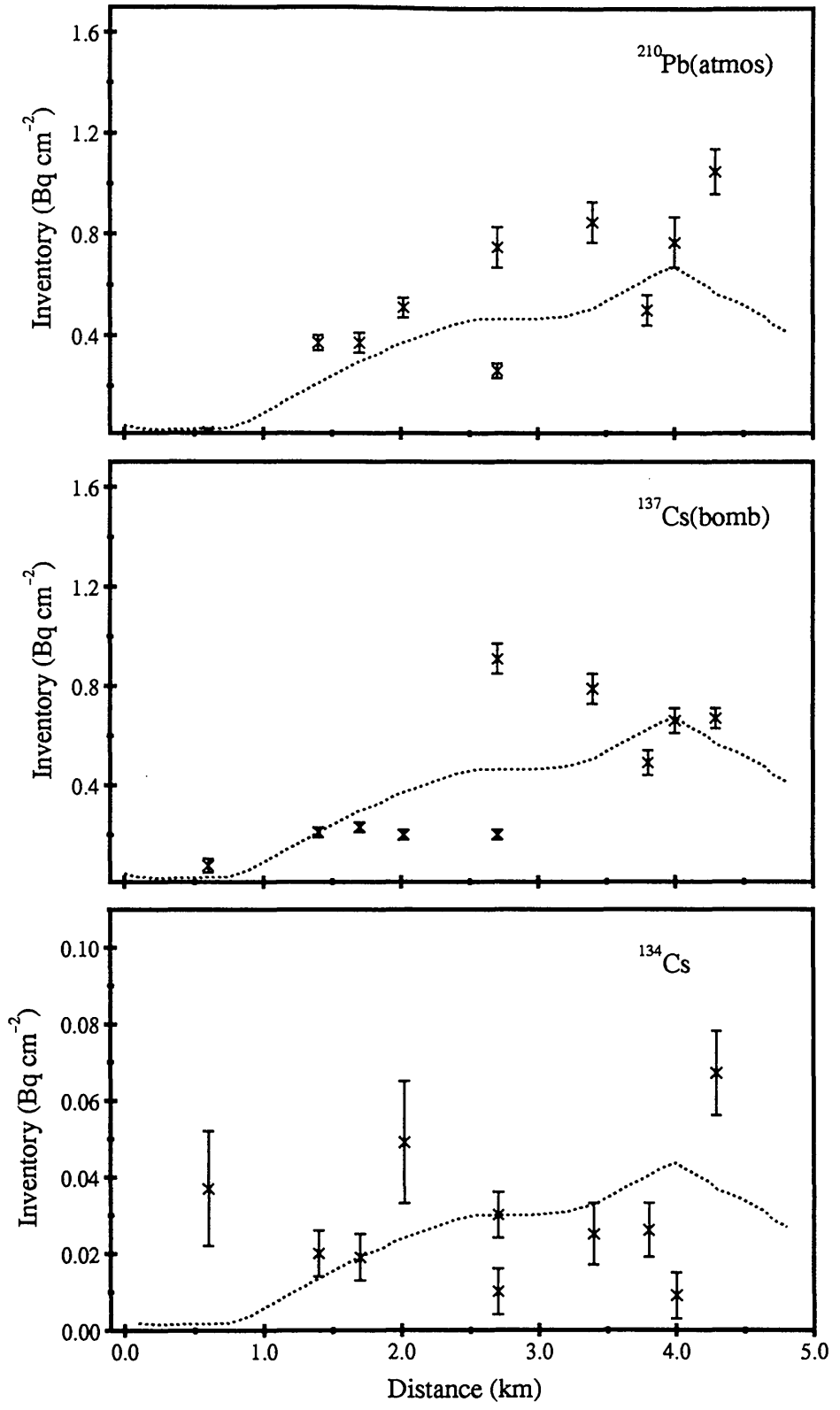

Figure 4.16: ^{210}Pb(atmos), ^{137}Cs(bomb) and ^{134}Cs inventories measured at Ben Cruachan

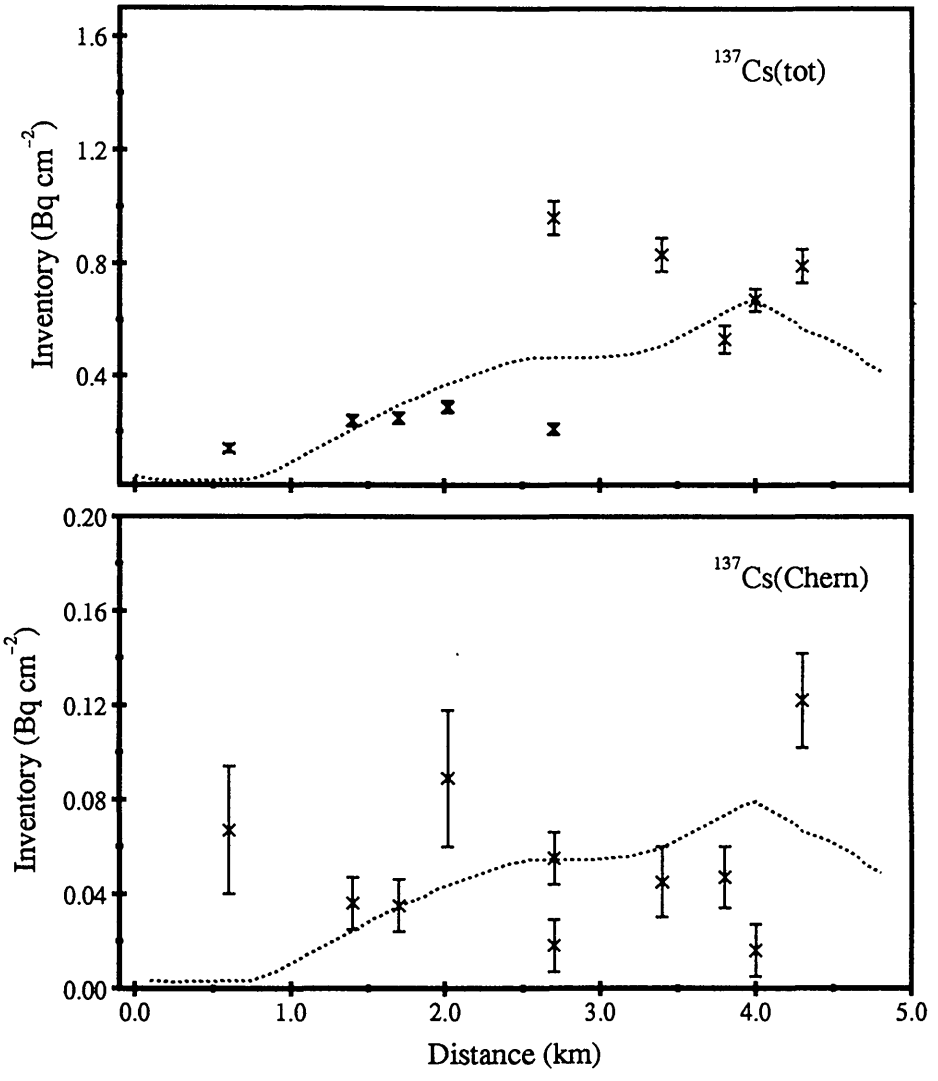

Figure 4.17: ^{137}Cs(tot) and ^{137}Cs(Chern) inventories measured at Ben Cruachan

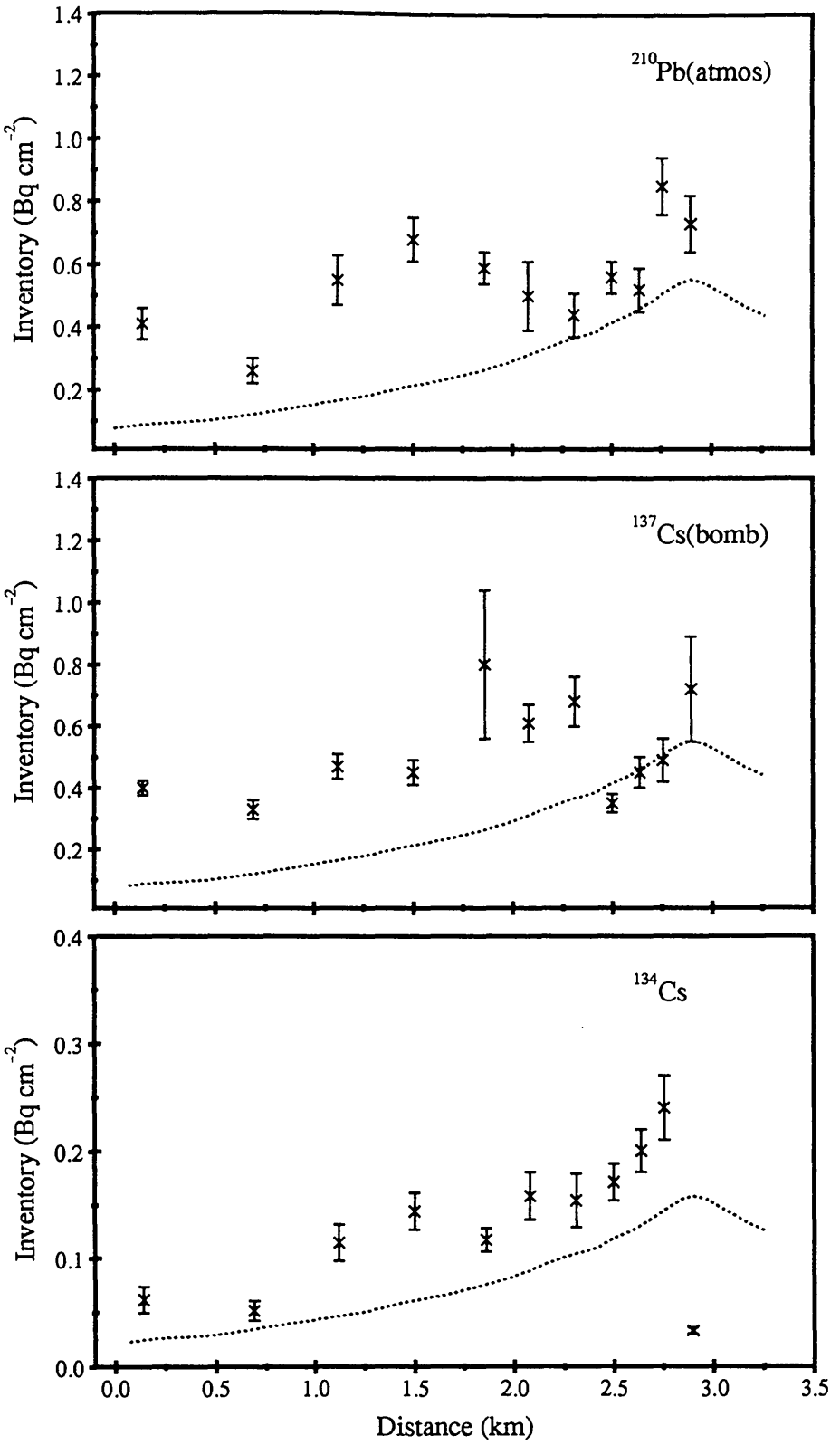

Figure 4.18: ^{210}Pb(atmos), ^{137}Cs(bomb) and ^{134}Cs inventories measured at Beinn Dorain

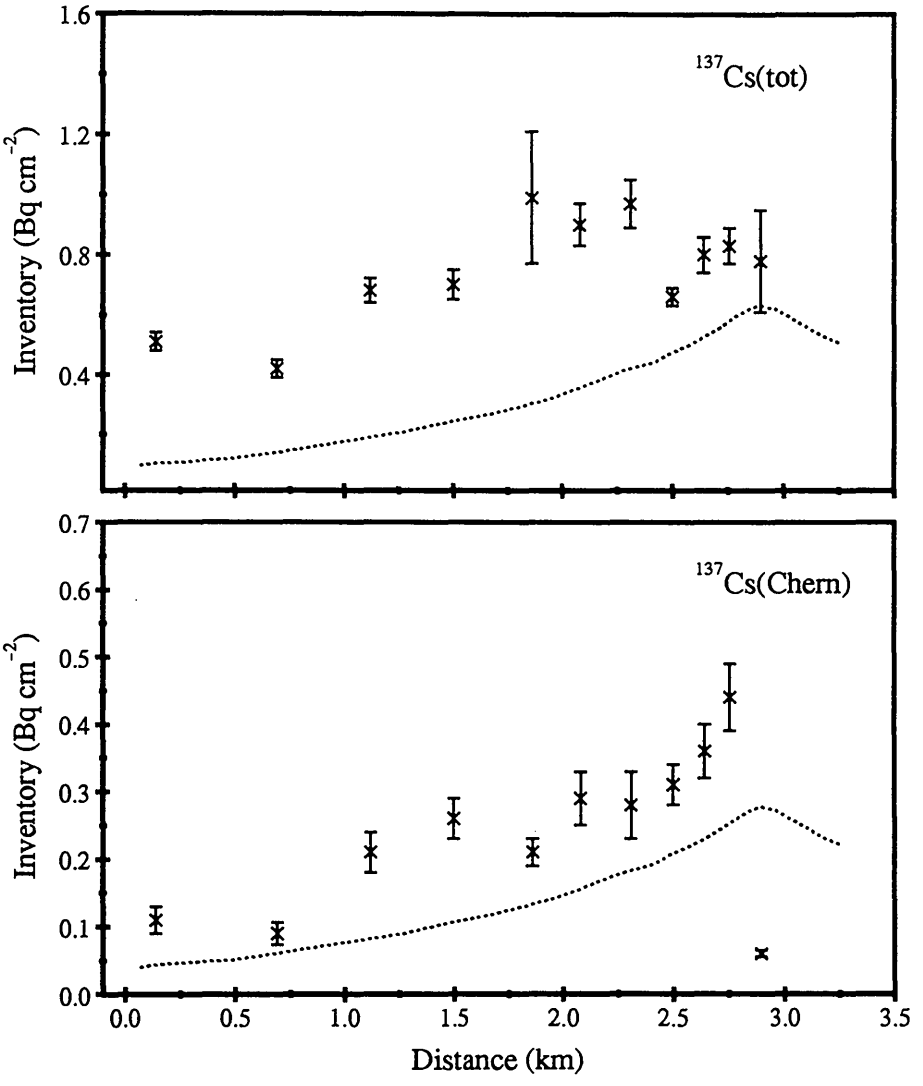

Figure 4.19: ^{137}Cs(tot) and ^{137}Cs(Chern) inventories measured at Beinn Dorain

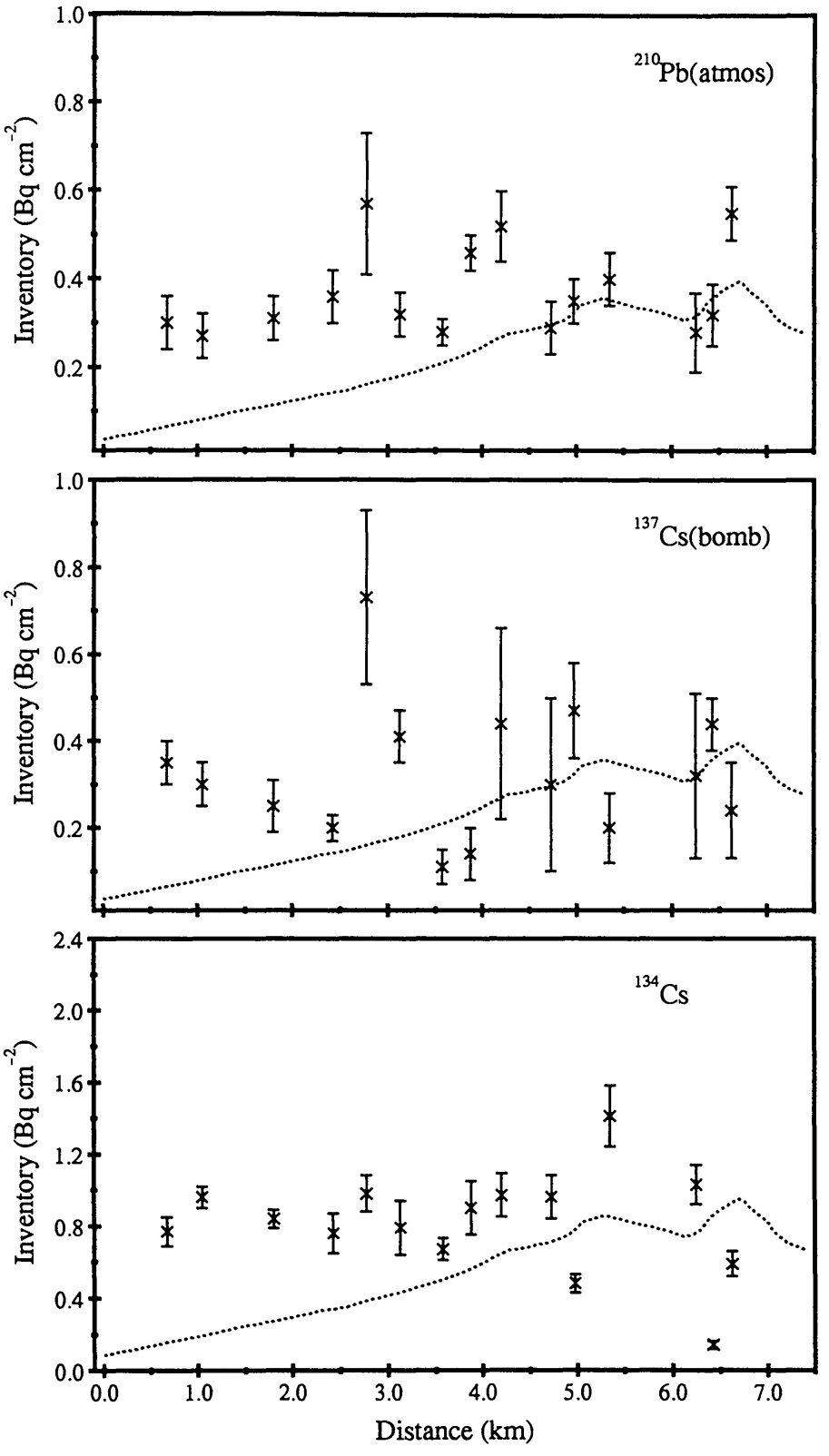

Figure 4.20: ^{210}Pb(atmos), ^{137}Cs(bomb) and ^{134}Cs inventories measured at Ben Lawers

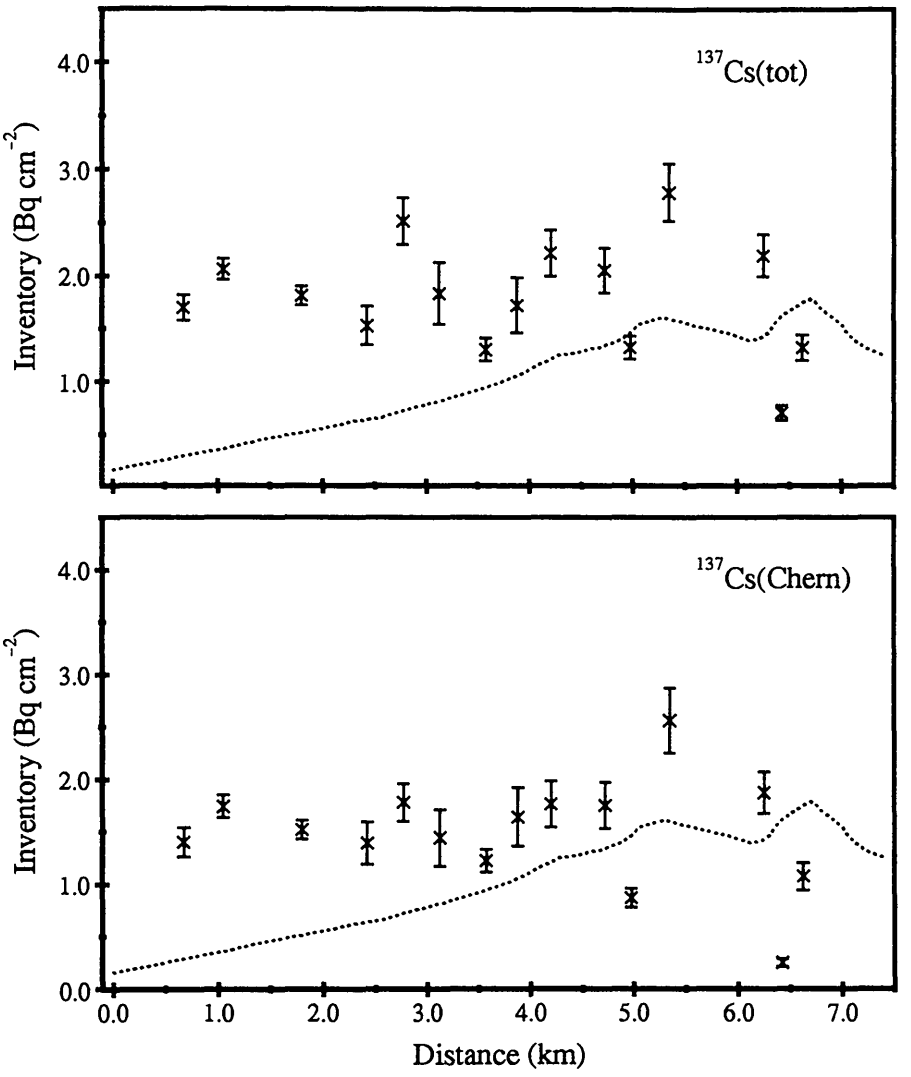

Figure 4.21: ^{137}Cs(tot) and ^{134}Cs inventories measured at Ben Lawers

4.2 Loss-on-ignition Results

As the composition of the soil in terms of mineral content and organic matter content influences the retention of ^{210}Pb and Cs isotopes, loss-on-ignition (LOI) experiments were carried out on certain samples. The samples had already been oven-dried in preparation for γ-ray analysis. The results of the LOI experiments are presented in Tables 4.1 to 4.4. The tables show the percentage of weight lost from each depth interval after heating the samples for 12 hours at about 500°C. The uncertainties associated with the core values in Table 4.2 represent the standard deviations of 3 replicates. Table 4.1 shows the results for each pit and set of cores from the Merrick sites M1–6. Table 4.3 shows the results from one of the cores taken from site M7 and one taken from site M8.

Depth interval	Loss-on-ignition (%)					
	M1	M2	M3	M4	M5	M6
0–5 cm	53	43	76	58	30	14
5–10 cm	33	23	38	28	21	9
10–20 cm	23	20	24	19	19	5
20–25 cm	–	14	–	–	–	5

Table 4.1: Results of the loss-on-ignition experiments carried out on the pit samples collected from sites M1–M6. Values represent the percentage of weight lost by the sample after being placed in an oven at 500 °C

At each of the 8 sites a decrease in the organic matter content is observed with depth down the soil profile. Considering sites M1–M6 it appears that sites M1–M4 show similar organic matter characteristics (except maybe for the highly organic pit sample from site M3). The organic matter content at M6 is much lower than for sites M1 to M4. Site M5 shows characteristics which seem to be intermediate between on the one hand, sites M1–M4 and, on the other, site M6. The values

Depth interval	Loss-on-ignition (%)					
	M1	M2	M3	M4	M5	M6
0–1 cm	84±2	73±4	86±5	81±4	55±4	21±1
1–10 cm	38±9	30±4	38±3	33±2	26±7	12±2

Table 4.2: Results of the loss-on-ignition experiments carried out on the core samples collected from sites M1–M6

Site M7, point (i)		Site M8, point (i)	
Depth interval	LOI (%)	Depth interval	LOI (%)
0–1 cm	83	0–1 cm	95
1–5 cm	26	1–5 cm	94
5–7.5 cm	12	5–10 cm	89
7.5–10 cm	11	10–15 cm	88
10–15 cm	10	15–20 cm	54

Table 4.3: Results of the loss-on-ignition experiments carried out on samples collected from sites M7 and M8

obtained from the LOI experiments are sensitive to the depth interval which the sample represents. As a result it is difficult to compare directly the results from sites M7 and M8 with those from sites M1 to M6. However, the 0–1 cm organic matter content at site M7 is more similar to those at sites M1 to M4 than those at M5 or M6. At site M7 there is quite a rapid fall off of organic matter content with depth down the profile. Site M8 is extremely organic, the site with by far the highest organic matter content which, even at 15–20 cm depth, comprises 54% organic matter by weight.

Table 4.4 gives the loss on ignition for the Ben Cruachan sites C5 and C9. The two sets of cores from site C5 were analysed separately since they gave such different values for the total inventory of ^{210}Pb(atmos) and ^{137}Cs(bomb) at this site and it was thought that this difference could arise from differences in the organic matter content of the two sampling locations. Samples from site C9 were analysed since the measured inventories of ^{210}Pb(atmos) and ^{137}Cs(bomb) at this site were extremely low.

The results from the two sampling points at site C5 and from the sampling point at site C9 show a decreasing proportion of organic matter with depth. At site C5 sampling point (i) is more organic in content point (ii). The organic matter content at site C9 is low.

Depth interval	Loss-on-ignition (%)		
	Site C5		Site C9
	point (i)	point (ii)	
0–1 cm	91	72	13
1–5 cm	65	45	8
5–10 cm	58	20	5
10–15 cm	46	36	–

Table 4.4: Results of the loss-on-ignition experiments carried out on samples collected from sites C5 and C9

4.3 Mean Annual Rainfall

Over the British Isles the rainfall amount reaching the ground increases with altitude. The removal of ^{210}Pb and Cs isotopes from the atmosphere occurs predominantly by the process of wet deposition and it is to be expected that the inventories of these radionuclides in the soil will be closely linked to the rainfall pattern. The UK rainfall field is obtained from the measurements of $\sim$4,000 collectors but few are located at altitude and at none of the mountains in this study have there been detailed measurements of the variation in rainfall with altitude. The UK rainfall field has been produced by interpolation between the collector sites with adjustments made for the effects of orography. Profiles of the variation in precipitation over each of the 5 mountains in this study are presented in Figures 4.22 and 4.23. The source of these profiles is the mean annual rainfall map of Britain [107]. The rainfall values at low altitudes are probably accurate to $\sim$10% whereas at higher altitudes the uncertainty in the values could be of the order of 30%. Table 4.5 gives the factor increase in the rainfall amount between the valley, to the west or southwest, and the summit for each mountain location.

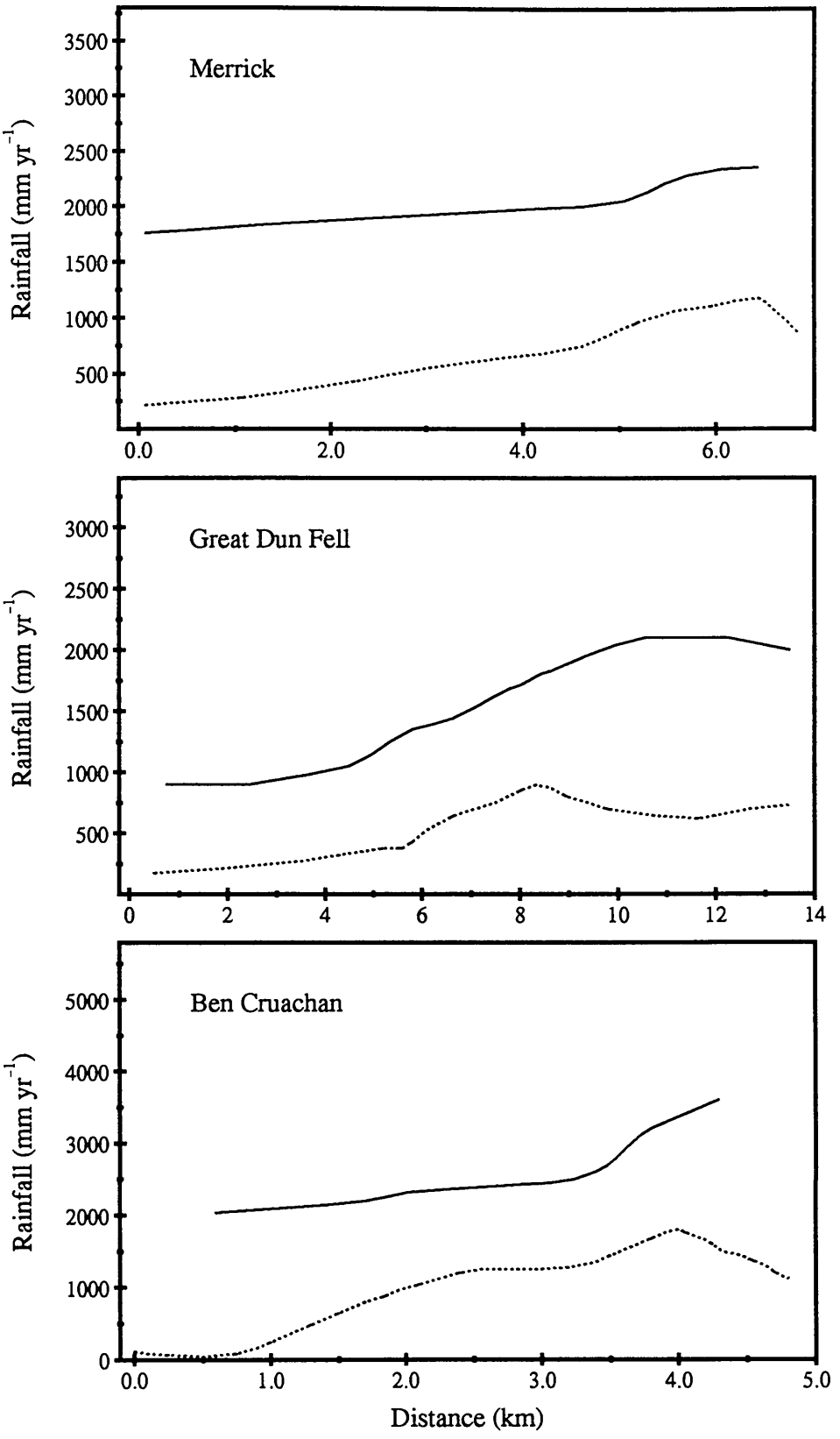

Figure 4.22: Mean annual rainfall (solid line) taken from the 30 year average annual rainfall map [107]. The dotted line represents the profile of the hill

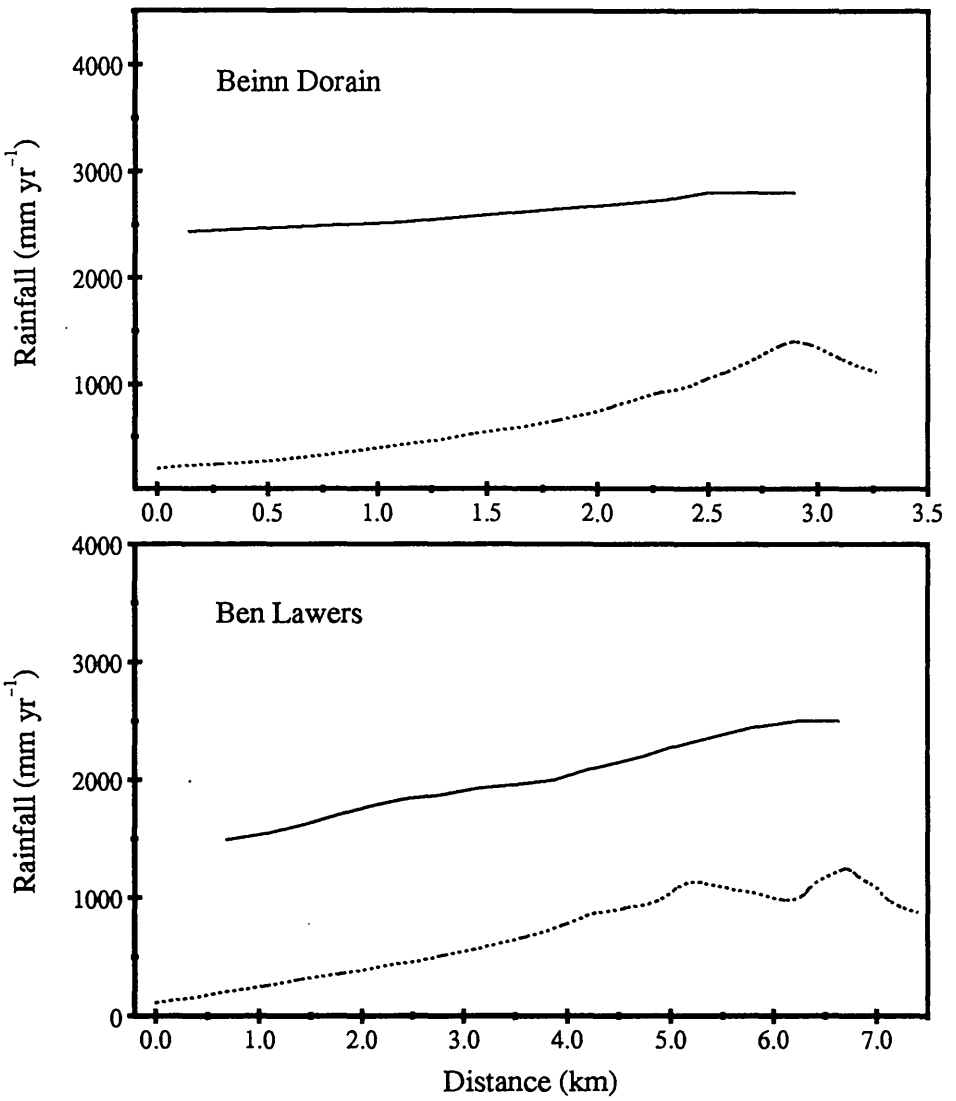

Figure 4.23: Mean annual rainfall (solid line) taken from the 30 year average annual rainfall map [107]. The dotted line represents the profile of the hill

Mountain location	Factor increase in average rainfall
Merrick	1.3
Great Dun Fell	2.0
Ben Cruachan	1.7
Beinn Dorain	1.2
Ben Lawers	1.7

Table 4.5: Factor increase in the average annual rainfall between the summit of each mountain and the valley to the west or southwest. Data taken from the 30 year (1941–70) average annual rainfall map [107]

Chapter 5

Discussion of the ^{210}Pb results

5.1 The profile of the atmospheric ^{210}Pb Inventory at each mountain

Table 5.1 lists some important characteristics of each of the 5 mountains sampled in this study. These are; altitude range of sampling, distance from the western coast, mean height 20 km upwind, gradient of the slope, half-length of the mountain, increase in rainfall between the valley and the summit and the corresponding increase in the ^{210}Pb(atmos) inventory. In Figures 5.1 and 5.2 the inventory of ^{210}Pb(atmos) at each mountain is shown. At four of the five mountains the evidence suggests that the ^{210}Pb(atmos) inventory increases with altitude. The exception is at Ben Lawers where there is no clear increase with altitude. Six values have been removed from the plots in order to simplify the interpretation of the measured ^{210}Pb(atmos) profiles. These six values represent the sites, M7, M8, G8, C3, C9 and sampling point (i) at site C5. In sections 5.1.1 to 5.1.5 the results from each mountain are considered in turn, and the reasons are given for the exclusion of these six points from the analysis of the deposition profiles.

Feature	Merrick	Great Dun Fell	Ben Cruachan	Beinn Dorain	Ben Lawers
Altitude range of sampling (m)	150–843	160–840	50–1,126	210–1,074	200–1,190
Distance from west coast (km)	30	70	30	55	80
Mean height 20 km upwind (m)	180	200	170	380	680
Gradient	1:9	1:9	1:4	1:3	1:6
Half length of hill (km)	2.7	2.5	2.2	1.0	2.0
Rainfall increase between valley and summit	1.3	2	1.7	1.2	1.7
^{210}Pb(atmos) inventory increase between valley and summit	2.5	3.3	2.8	2.4	<1.8

Table 5.1: Important characteristics of each of the mountains sampled in this study

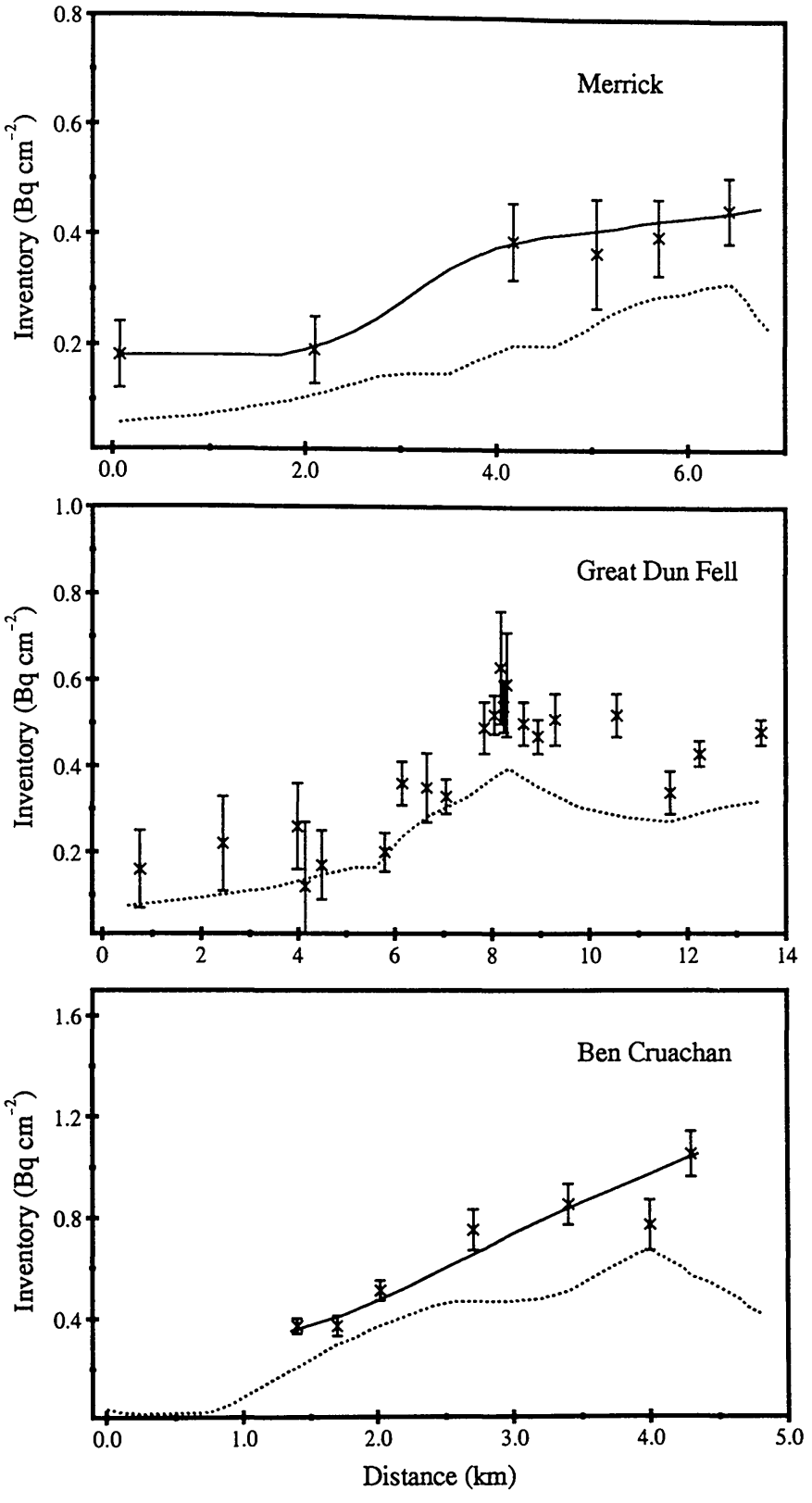

Figure 5.1: Inventory of ^{210}Pb(atmos) (Bq cm^{-2}) at the Merrick, Great Dun Fell and Ben Cruachan

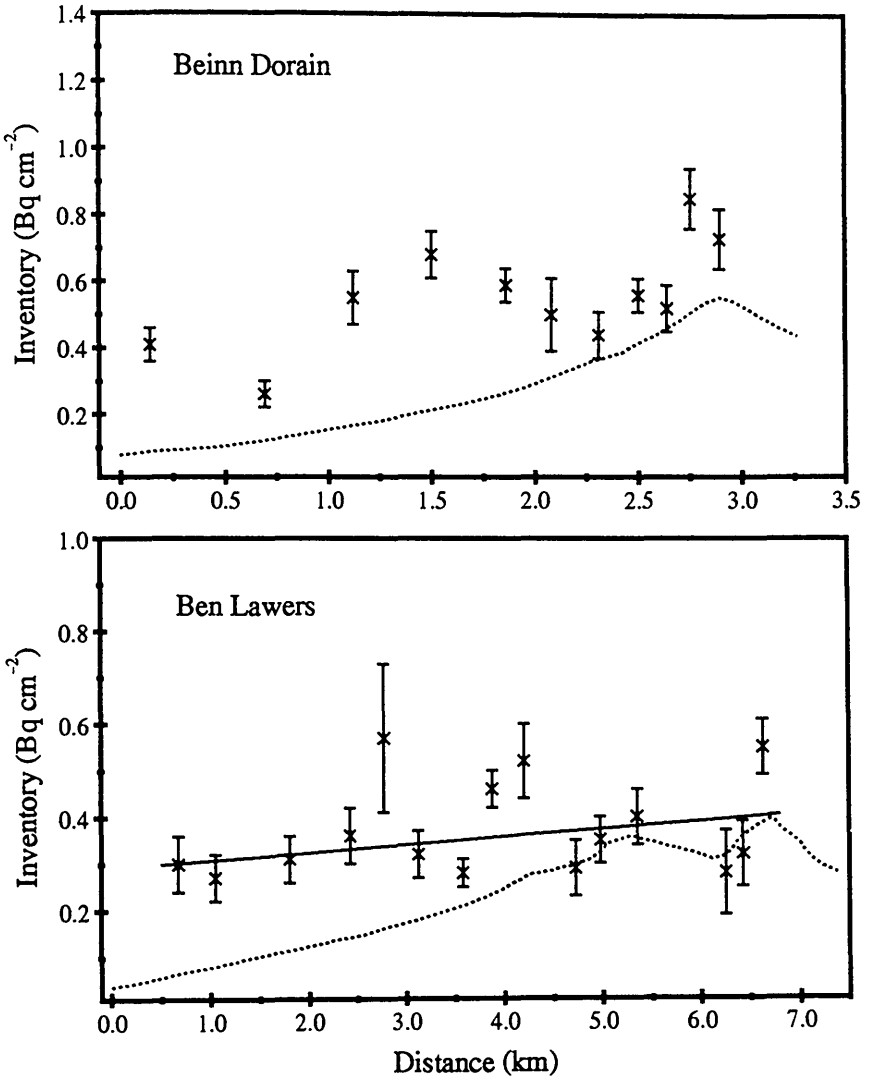

Figure 5.2: Inventory of ^{210}Pb(atmos) (Bq cm^{-2}) at Beinn Dorain and Ben Lawers

5.1.1 Merrick

Looking at Figures 4.1 and 4.3 it can be seen that the measurements of the ^{210}Pb(atmos) inventory for sites M1–M6 agree quite well for the pit and core samples. The only site where the error bars of each independent measurement do not overlap is for site M3, where the pit value appears to be low. Interestingly, although the pit at M3 appears to be deficient in ^{210}Pb(atmos), it has a very high organic matter content, see Table 4.1. A similar increase, by a factor of 2–3, in the ^{210}Pb(atmos) inventory between the valley and the summit, is observed from the pits and from the cores. As can be seen in Figure 4.3, the results from sites M7 and M8 sampled at a later date than sites M1–M6 pose a problem. The ^{210}Pb(atmos) inventory values are roughly twice the size of the values from sites M5 and M6. There are several possible explanations to account for this result.

It is possible that sites M7 and M8 may be receiving inputs of ^{210}Pb on top of those being delivered directly from the atmosphere, for instance due to overland flow. Conversely sites M5 and M6 may be 'losing' their atmospherically derived ^{210}Pb(atmos), for example due to a lack of organic matter to trap the ^{210}Pb(atmos), or due to physical erosion of organic particles. The environment of sampling site M5 was a gently sloping grassy patch within an area of moorland, M6 was a flat pasture site a few metres away from a small river, M7 was a gently sloping pasture site and M8 was on the side of a sloping bank, just above an area of marsh. Site M6 possesses characteristics which suggest that it may not be retaining ^{210}Pb(atmos), specifically its proximity to a stream and the fact that it is deficient in organic matter, see Table 4.1. However, the values from this site are in line with those from site M5 and the soil at site M6 is not likely to be any wetter than the soil at site M8. It is difficult to ascribe the differences in the results from sites M5 and M6 and those from sites M7 and M8 to site characteristics.

An alternative explanation is that the differences are largely due to sampling procedure and variation in the detector performance. Cores taken from sites M7 and M8 were subject to finer depth division than the core and pit samples

from sites M1–M6. The low branching ratio (4.05%) of the ^{210}Pb γ-ray, self-absorbtion within the sample itself, the relatively high background of the γ-ray spectrum at low energy and the fact that the bulk of the ^{210}Pb(atmos) is retained within a few cm of the surface means that counting may be sensitive to dilution of the ^{210}Pb(atmos) within a sample which covers a large depth interval. Also the performance of the 19% Ge(Li) detector, in terms of resolution, was improved significantly between the counting of the samples from sites M1–M6 and those from M7 and M8. This factor may have aided the measurement of low, but significant, concentrations of ^{210}Pb(atmos) in the later samples. This factor, coupled with the different sampling procedure, is thought to be the cause of the conflicting results between sites M5,M6 and sites M7,M8. In this situation, it is thought that there is a systematic shift in the measured values between those from sites M1–M6 and those from M7 and M8.

This conclusion has important implications for planned sampling expeditions, to ensure a consistent sampling procedure, and ensuring detector system stability during analysis of samples from the same location. The work at this location was the pilot study for subsequent work. Lessons were learned here, and the ideas discussed above were used in later sampling and analysis. The interpretation of the Merrick results will focus on the values obtained from sites M1–M6. However, the results from sites M7 and M8 show that the conclusions should be treated with caution.

Three sets of cores were taken from sites M1–M6. The mean coefficient of variation for these 6 sites is 34% with values ranging from 8% to 94% (at one sampling site). As the results for the pit and core samples from sites M1–M6 are so similar, they have been combined in Figure 5.1. The solid line indicates the inferred inventory of ^{210}Pb(atmos) over the hill profile. The value in the valley, at site M6, is 0.18 Bq cm^{-2} and at the summit, site M1, it is 0.45 Bq cm^{-2}. Thus, the inventory at the summit is a factor of 2.5 greater than that in the valley, this compares with a factor increase in rainfall of $\sim$1.3 between the valley and the summit.

The Merrick is situated $\sim 30\,\mathrm{km}$ from the coast and the rainfall is about $1{,}800\,\mathrm{mm\,yr^{-1}}$ at the lowest site. This is large compared to the coastal rainfall which is, typically, $\sim 1{,}000\,\mathrm{mm\,yr^{-1}}$ at the western coast. (The lowest sites at Great Dun Fell receive $\sim 900\,\mathrm{mm\,yr^{-1}}$.) It seems that the rainfall at the base of the Merrick is already enhanced due to uplifting of airmasses by the extent of land, of mean height 200 m asl, between the Merrick and the coast, see Figure 3.2. The ^{210}Pb(atmos) inventory which would be expected at the coast may be estimated by extrapolating from the value at site M6. This gives a ^{210}Pb(atmos) inventory of $0.10 \pm 0.04\,\mathrm{Bq\,cm^{-2}}$ with $1{,}000\,\mathrm{mm\,yr^{-1}}$ rainfall at the coast. At the summit of the Merrick the measured ^{210}Pb(atmos) inventory is $0.45 \pm 0.06\,\mathrm{Bq\,cm^{-2}}$ with $2{,}400\,\mathrm{mm\,yr^{-1}}$ of rainfall. In addition, occult deposition of cloud water is not measured by conventional raingauges, and thus it is likely that the amount of water deposited at the summit is greater than is indicated on the rainfall map. Fowler et al. [59] measured cloud water deposition at a site at 620 m asl at Great Dun Fell and calculated an input of $116\,\mathrm{mm\,yr^{-1}}$ cloud water to moorland vegetation. Rainfall at this altitude at Great Dun Fell is $1{,}440\,\mathrm{mm\,yr^{-1}}$; thus the input due to occult deposition amounts to 8% of the rainfall total. If, say, occult deposition is responsible for an extra 10% of water at the summit of the Merrick (ie 10% of the $2{,}400\,\mathrm{mm\,yr^{-1}}$ indicated on the average annual rainfall map) then the input of liquid water becomes $2{,}640\,\mathrm{mm\,yr^{-1}}$.

These figures represent a factor increase, between the summit and the coast, of 4.5 in the ^{210}Pb(atmos) inventory, and 2.64 in the annual liquid water input. This corresponds to a concentration of ^{210}Pb in water input to the summit $\times 1.7$ (± 0.7) greater than that in seeder rain falling at the coast. If the increase in the ^{210}Pb(atmos) inventory and liquid water input, between the coast and the summit, is due solely to removal of cloud droplets from low level air, then the mean concentration of ^{210}Pb in the cloud droplets is $\times 2.1$ (± 0.8) greater than in the seeder rain at the coast. It appears that the enhancement at the Merrick takes place above 300 m asl although it is difficult to locate the altitude at which it becomes detectable from 6 sampling sites.

5.1.2 Great Dun Fell

The ridge of hills, of which Great Dun Fell is one element, runs in a northwest to southeast direction and the individual mountains rise to altitudes of up to 900 m asl. Most of the rainfall falling onto Great Dun Fell is due to frontal systems originating from the west and southwest. As a result, southwest of the summit will be referred to as 'upwind' of the summit and northeast of the summit will be referred to as 'downwind'.

The values of the ^{210}Pb(atmos) inventory at Great Dun Fell are shown in Figure 5.1. The main characteristics of the distribution are that the values are roughly constant in the Eden Valley, although the error bars are large, and that the values begin to rise sharply at the break in slope, at about 5.5 km distance along the transect. The largest values are found at the summit, although, downwind of the summit values remain greater than those in the Eden Valley. The wooded site, G8, produced a value of the ^{210}Pb(atmos) inventory of $0.41 \pm 0.13\,\mathrm{Bq\,cm^{-2}}$, which is twice the size of the mean baseline value for the Eden Valley. Though the error bars, on the value from this site, do overlap with the values from the neighbouring sites, G9 and G10, see Figure 4.14.

In order to assess the magnitude of the increase in the ^{210}Pb(atmos) inventory with altitude at Great Dun Fell a baseline value for the Eden Valley was obtained by taking the mean of the six sites; G6, 7, 9, 10, 11, 16. The mean of these 6 sites has a value of $0.19 \pm 0.04\,\mathrm{Bq\,cm^{-2}}$ and the corresponding mean value of the average annual rainfall is $1{,}040\,\mathrm{mm\,yr^{-1}}$.

Before considering the enhancement of the ^{210}Pb(atmos) inventory with altitude at Great Dun Fell it is useful to discuss the deposition pattern. Of the 23 sites, the four highest values of the ^{210}Pb(atmos) inventory are found at sites G1, 2, 3 and 12 on the western, 'upwind' slopes of Great Dun Fell, within 500 m distance of the summit. Models of seeder-feeder scavenging predict that for a hill like Great Dun Fell the maximum rainfall rate and maximum deposition rate of chemical species is downwind of the summit when there is a cap cloud present, in

contact with the hill surface, and rain bearing frontal systems coming from a west or southwesterly direction [24][80]. The shift in the zone of maximum deposition downwind of the summit at 'short' hills (ie those with a half length of $\sim 2\,\mathrm{km}$), such as Great Dun Fell, is due to the wind-drift of the raindrops falling from the seeder-cloud. The zone of highest liquid water content in the feeder-cloud is centred over the summit, where the cloud droplets are largest. Because of the wind-drift of the seeder raindrops those drops which have the longest path length through the zone of high liquid water content in the feeder-cloud will reach the ground surface downwind of the summit. Maximum deposition at Great Dun Fell is therefore expected to be 1–2 km downwind of the summit.

However, the results of this study indicate the maximum deposition of ^{210}Pb to be at, and just upwind, of the summit. Two possible explanations of the observed profile of ^{210}Pb(atmos) over Great Dun Fell will be discussed.

The first explanation of the observations is that seeder-feeder scavenging at Great Dun Fell results in a maximum of deposition at and just upwind of the summit. Hill et al. [80] ran their model of seeder-feeder scavenging with a set of standard conditions, typical of values measured at Great Dun Fell, as the input. These standard conditions include a supercritical airflow regime (characteristic of hills whose height is comparable to the boundary layer depth), geostrophic wind of $15\,\mathrm{m\,s^{-1}}$, hill height of 665 m, height of cloud base 300 m, rainfall rate $1\,\mathrm{mm\,h^{-1}}$ and depth of feeder-cloud 800 m. Running the model with the standard input parameters the maximum precipitation rate is seen to be downwind of the summit. However, when a lower windspeed of $5\,\mathrm{m\,s^{-1}}$ is input, the maximum rate of precipitation is observed upwind of the summit.

The model of Hill et al. also predicts the deposition rate, and the form of the deposition pattern, for chemical species. With standard input parameters the maximum deposition of sulphate is seen to lie downwind of the summit. Yet, in the case when a higher rainfall rate of $5\,\mathrm{mm\,h^{-1}}$ is input, and also in the case when the thickness of the feeder cloud is halved to 400 m, then the zone of maximum sulphate deposition is seen to be centred around the summit. Thus, under

certain conditions, the seeder-feeder scavenging model predicts a zone of maximum precipitation and maximum sulphate deposition at Great Dun Fell which is not downwind of the summit but centred around the summit. The deposition profile inferred from the measurements, by this argument, is shown in Figure 5.3. However, it must be said that the parameters needed to produce a maximum of precipitation and sulphate deposition upwind of and around the summit are not those typical, and most frequently observed, of the conditions at Great Dun Fell. It is also difficult to conceive how the seeder-feeder scavenging process operating at this mountain could result in a maximum of ^{210}Pb deposition around the summit when the maximum annual precipitation is 2 km downwind of the summit (see Fig 3.22).

The second explanation is that the zone of maximum deposition due to seeder-feeder scavenging is downwind of the summit and that high rates of occult deposition around the summit lead to the maximum of deposition, due to all processes, being located at the summit itself. Mean windspeeds are frequently greatest at and just upwind of the hill summit. Assuming that the surface roughness over Great Dun Fell is constant, then turbulence will be greatest where the highest windspeeds are found. Thus, turbulent deposition of cloud droplets is expected to be a maximum at and just upwind of the hill summit. The turbulent deposition model of Hill et al. [80] predicts that the maximum rates of cloud water deposition will be located at the summit of Great Dun Fell. An estimate can be made for the contribution of occult deposition to the total deposition of ^{210}Pb at the summit. The mean ^{210}Pb(atmos) inventory from the four highest value sites at the summit, sites G1, 2, 3 and 12, is 0.58 ± 0.05 Bq cm^{-2}. The mean ^{210}Pb(atmos) inventory from the two sites just downwind of the summit, sites G17 and 18, is 0.49 ± 0.03 Bq cm^{-2}. The assumptions are made that sites G17 and 18 are sheltered from the very high occult deposition rates at and just downwind of the summit and that, at these sites, occult deposition accounts for 10% of total deposition of ^{210}Pb(atmos). An estimate for the contribution of occult deposition to total deposition of 10% is reasonable for a high altitude moorland site [59]. Two

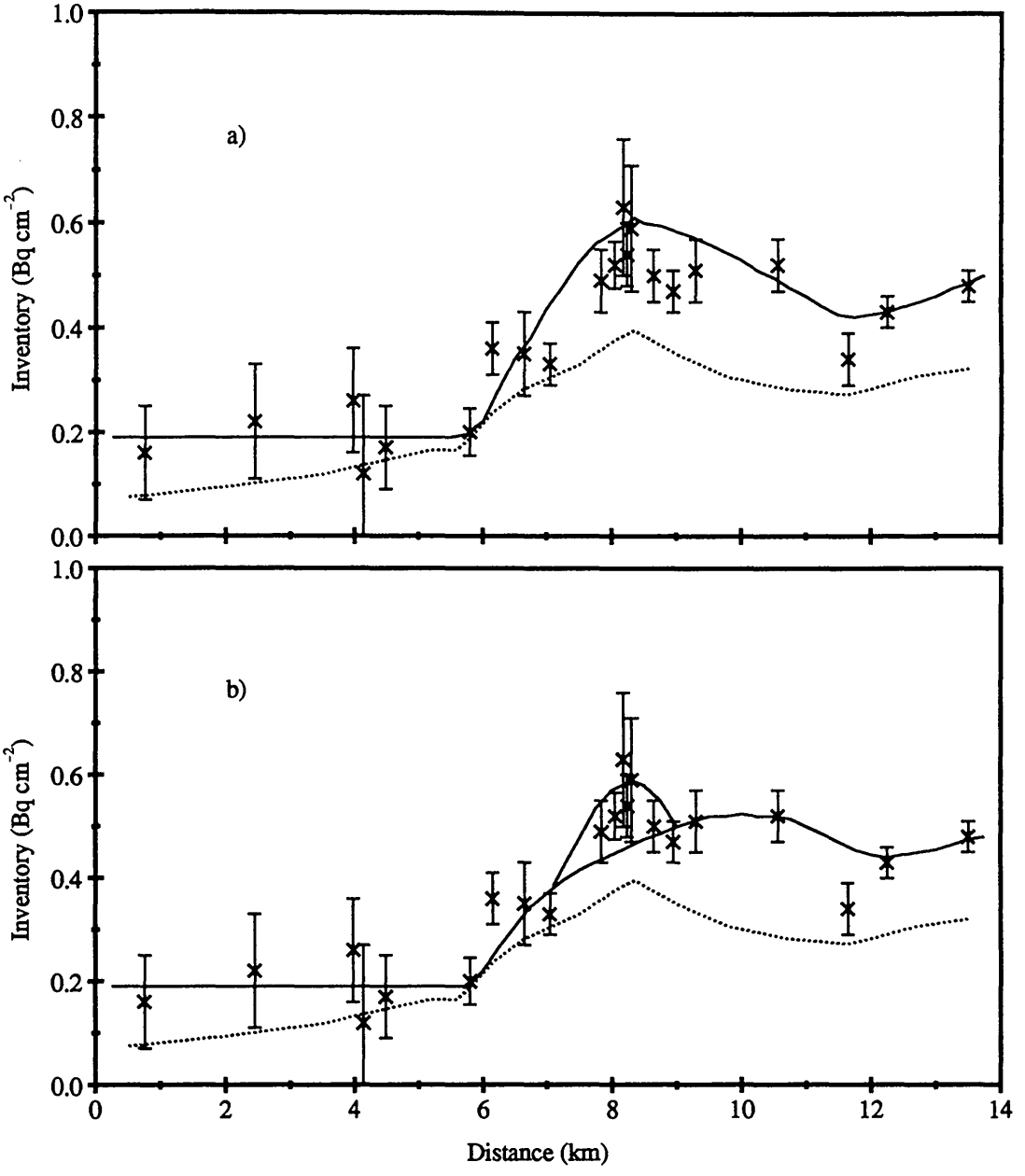

Figure 5.3: The two interpretations of the measured ^{210}Pb(atmos) profile over Great Dun Fell. The dotted line represents the profile of the hill and the solid line represents the profile inferred by the measured points

additional assumptions are made; that the contribution of dry deposition to total deposition is negligible and that the deposition of ^{210}Pb in rainfall at the downwind sites is the same as that at the summit sites. Given the short horizontal distances between the sites, this assumption is not unreasonable. Following this reasoning then, at the downwind sites occult deposition accounts for $\sim 0.05\,\mathrm{Bq\,cm^{-2}}$ of the ^{210}Pb(atmos) inventory and wet deposition for $\sim 0.44\,\mathrm{Bq\,cm^{-2}}$. Assuming the wet deposition contribution is the same at the summit and just downwind, then occult deposition contributes $\sim 0.14\,\mathrm{Bq\,cm^{-2}}$ to the ^{210}Pb(atmos) inventory at the summit. This figure amounts to 23% of the total deposition of ^{210}Pb and 30% of the amount deposited in rainfall. This is higher than the figure given by Dollard et al. [45]. They found that, on average, occult deposition to moorland at Great Dun Fell, increases the measured wet deposition amounts by 20%. The interpretation that occult deposition is greatest at the summit is in contrast with the model of Hill et al. [80] which predicts that the turbulent deposition of sulphate is a maximum downwind of the hill summit.

There is good evidence that the maximum in wet deposition is displaced to the northeast of the summit if we accept the conclusion that the values at the summit are enhanced by occult deposition whereas the sites downwind of the summit are not so greatly affected. The inferred deposition in Figure 5.3 shows that values may increase slightly downwind of the summit. Of all the sites downwind of the summit it is sites G19 and G20 at, respectively, 1 and 2 km downwind of the summit which give the greatest values of the ^{210}Pb(atmos) inventory. Comparing sites at similar altitude on the western and eastern slopes of the mountain, the sites G13 and 14 (at 650 m and 600 m asl respectively) to the west have a mean ^{210}Pb(atmos) inventory of $0.34 \pm 0.04\,\mathrm{Bq\,cm^{-2}}$ while sites G19 and G20 (of altitude 700 m and 600 m respectively) to the east have a mean inventory of $0.52 \pm 0.04\,\mathrm{Bq\,cm^{-2}}$. Thus, the ^{210}Pb(atmos) inventory is a factor of 1.5 greater at the same altitude to the west than to the east of the summit. This difference reflects the difference in rainfall amount as the mean of the average annual rainfall for the eastern sites is a factor of 1.4 greater than that for the western sites. The

results indicate that the wind drift of seeder rain drops falling through the feeder cloud displaces the maximum of precipitation amount and deposition of ^{210}Pb in rain to the northeast of the summit.

In view of the preceding discussion, the second explanation of the measured ^{210}Pb(atmos) profile is the more likely. This is, that seeder-feeder scavenging enhances rainfall amount and the concentration of ^{210}Pb in rainfall, and has a maximum effect on the deposition downwind of the summit, while the maximum rate of occult deposition is located at the summit. The two processes combine to produce the observed deposition profile, as shown in Figure 5.3.

It is useful to calculate the enhancement of ^{210}Pb concentration in rainwater and cloudwater at high altitudes by considering the magnitude of the ^{210}Pb(atmos) variation with altitude together with rainfall and estimates of the input of water due to occult deposition. The baseline values calculated for the Eden Valley sites are $0.19 \pm 0.04\,\mathrm{Bq\,cm^{-2}}$ and $1{,}040\,\mathrm{mm\,yr^{-1}}$ for the ^{210}Pb(atmos) inventory and mean annual rainfall respectively. Gallagher et al. [63] estimate an input of water due to occult deposition of between 50–130 $\mathrm{mm\,yr^{-1}}$ to moorland vegetation at Great Dun Fell.

Downwind of the summit sites G19 and 20 give a mean ^{210}Pb(atmos) inventory of $0.52 \pm 0.04\,\mathrm{Bq\,cm^{-2}}$ and rainfall of $2{,}020\,\mathrm{mm\,yr^{-1}}$. An input of liquid water due to occult deposition of $50\,\mathrm{mm\,yr^{-1}}$ is assumed (ie Gallagher et al.'s lower value) giving a liquid water input of $2{,}070\,\mathrm{mm\,yr^{-1}}$ at these sites. Using these figures the enhancement of the ^{210}Pb(atmos) inventory is a factor of ~ 2.7. The enhancement in the input of liquid water is a factor of ~ 2.0. This result points to the concentration of ^{210}Pb in the liquid water deposited downwind of the summit, (ie rainfall and occult precipitation combined) being a factor of 1.4 ± 0.4 greater than that in the rainfall deposited in the valley (ie the seeder rainfall). The concentration of ^{210}Pb in cloud water is indicated to be on average 1.8 ± 0.5 greater than that in the seeder rain.

At the summit, sites G1, 2, 3, 12 give a mean inventory of $0.58 \pm 0.05\,\mathrm{Bq\,cm^{-2}}$ and a mean yearly rainfall of $1{,}760\,\mathrm{Bq\,cm^{-2}}$. An occult precipitation input of

130 mm yr^{-1} is assumed (ie Gallagher et al.'s upper limit), giving a total yearly liquid water input of 1,890 mm yr^{-1}. The ^{210}Pb(atmos) inventory at the summit is a factor of ~ 3.1 greater than the mean value in the valley and the liquid water input is a factor of ~ 1.8 greater. This indicates that the concentration of ^{210}Pb in liquid water deposited to the summit is a factor of 1.7 ± 0.5 greater than that in the seeder rain. The concentration of ^{210}Pb in cloudwater is calculated to be a factor of 2.6 ± 0.7 greater than the seeder rainfall. These results calculated from the downwind and summit sites point to a mean concentration ~ 1.6 (± 0.3) times greater in water deposited at high altitudes than in the seeder rain and a concentration in the feeder cloud water which is on average a factor of ~ 2.2 (± 0.4) greater than that in seeder rain.

The results from the downwind and summit sites are the same within the error bars but the difference between the mean values may be significant. The difference could be due to the uncertainty in the estimates of the amount of water deposited to the ground surface by occult deposition, assuming the rainfall values are correct. The results could indicate that the difference between the amount of occult precipitation at the summit and downwind is greater than is estimated. Perhaps the estimate is too low for the summit sites or alternatively, at the summit, simultaneous turbulent deposition and evaporation from leaf surfaces may be taking place when thin cloud is present (ie < 0.1 g m^{-3} liquid water content). This would result in relatively high concentrations being deposited on leaf surfaces and subsequently washed to the ground by rainfall, Gallagher et al. [63].

It is interesting to consider the effect of a change in land use. If afforestation were to take place then the aerodynamic roughness would be increased from $z_o \simeq 2$ cm for rough grass to $z_o \simeq 50$ cm for forest, thus increasing the rates of turbulent deposition. The greater surface area presented by the forest would be expected to lead to high rates of impaction of cloud droplets at the forest edge. Fowler et al. [59] estimated that afforestation would increase the wet deposition input of major ions by $\sim 50\%$. They estimate that cloudwater deposition to moorland vegetation increases the wet deposition input by $\sim 12\%$. The turbulent

H^+	NH_4^+	Cl^-	NO_3^-	SO_4^{2-}	^{210}Pb
3.9	2.4	2.6	2.8	2.0	2.2

Table 5.2: Ratio of concentrations in cloud/rain for major ions and for ^{210}Pb at Great Dun Fell. Values for the major ions are the mean of 11 precipitation and cloud events during spring 1985

H^+	NH_4^+	Cl^-	NO_3^-	SO_4^{2-}	^{210}Pb
2.9	3.1	2.9	2.3	2.2	1.6

Table 5.3: Ratio of concentrations in rain, summit/valley, for major ions and for ^{210}Pb. Concentration ratios for the major ions were calculated from measurements made during 20 precipitation events at Great Dun Fell (1984–1985)

deposition model of Hill et al. [80] suggests that a forested hill has a turbulent deposition rate of sulphate 2.5–3 times higher than a grass covered hill. Afforestation would be expected to increase significantly the input of acidic ions to a hill like Great Dun Fell which is frequently shrouded in cloud.

The three sets of cores taken from site G8, beneath a small stand of conifers at 260 m altitude, give values of 0.46 ± 0.22, 0.36 ± 0.15 and $0.09 \pm 0.15\,Bq\,cm^{-2}$ for the ^{210}Pb(atmos) inventory. High variability in the ^{210}Pb(atmos) inventory beneath a forest is not unlikely, since drip of rain and cloud water from branches may fall unequally on the forest floor. Some locations may receive large amounts of water whereas others may be sheltered. Root disturbance of the soil may also take place at certain locations. The two high value sets of cores at site G8 give a mean inventory of $0.41 \pm 0.13\,Bq\,cm^{-2}$. This is twice the size of the baseline value for the Eden Valley, although the error bars overlap with those of the two adjacent sites G9 and 10, see Figure 4.14. This result could represent an increase in deposition of $\sim 100\%$ of wet deposition due to impaction and enhanced turbulent deposition of cloud droplets and large aerosols.

The results of this study indicate that, at Great Dun Fell, the flux of ^{210}Pb to the ground surface increases by a factor of ~ 3 and the mean concentration of ^{210}Pb in rainfall increases by a factor of ~ 1.6 between the Eden Valley and the summit. Low level cloud is estimated to contain a concentration of ^{210}Pb about twice that of seeder rain from above. Similar values have been obtained for the major ions in experiments carried out at Great Dun Fell and have been predicted in a model of seeder-feeder scavenging [32][57]. The results of Fowler et al. [57] are presented for comparison in Table 5.2 and Table 5.3 and represent measurements taken during autumn 1984 and spring 1985 while the airflow was from the west and southwest with a cap cloud and rainfall from above. The results of Fowler et al. were obtained during individual rainfall events under conditions favourable for the operation of the seeder-feeder mechanism. The ^{210}Pb(atmos) inventory records deposition under all meteorological conditions integrated over several decades. The fact that both studies produce comparable results indicates the importance of seeder-feeder scavenging and occult deposition to total deposition of chemical species at this mountain. The ^{210}Pb(atmos) results from Great Dun Fell give support to the ideas which point to the role of mountain cloud in delivering large quantities of acidic ions at high altitudes.

Climatic change would alter the deposition pattern of rainfall amount and acidic ions at Great Dun Fell. Observations indicate the importance of the seeder-feeder scavenging mechanism when airflow is from the west, but, when the airflow is from the east, no increase in concentration or rainfall amount is observed. The same is true when the winds are light or when flow over the hill is blocked in the Eden Valley [32]. A change in climate which caused a change in the annual rainfall amount would be expected to lead to an increase or decrease in the amount of wet deposition depending on whether the rainfall increased or decreased. At present the dominant airflow is from the west and southwest. If this were to switch to the east, then seeder-feeder scavenging would become much less important thus and the enhancement of acidic deposition with altitude would be reduced.

5.1.3 Ben Cruachan

Mean annual rainfall at Ben Cruachan is shown on the 30 year rainfall map, [107], to increase from 2,030 mm yr^{-1} in the valley at 50 m asl to 3,600 mm yr^{-1} to the east of the summit at 950 m asl. It is likely that, on the lower slopes of Ben Cruachan, some orographic enhancement of rain has been induced by the high ground to the southwest which rises up to $\sim$300 m asl. The mean annual rainfall along the western coast of Britain is typically $\sim$1,000 mm yr^{-1}.

The profile of ^{210}Pb(atmos) inferred by the measurements is shown in Figure 5.1. The inventory increases from 0.37 ± 0.03 Bq cm^{-2} at site C8 to a value of 1.05 ± 0.09 Bq cm^{-2} at site C2, to the east of the summit. A reliable value of the ^{210}Pb(atmos) inventory was not obtained for an altitude lower than that of site C8 at 350 m asl. Along the line chosen for sampling at this mountain, the ground level was very steep below the altitude of site C8. A low altitude site, C9 at 50 m asl, was selected which was level pastureland at the base of the mountain. In the event, though, this site yielded a very unlikely value for the atmospheric ^{210}Pb inventory. This site, C9, together with site C3 and point (i) at site C5 give values of the ^{210}Pb(atmos) inventory which are significantly lower than expected from the rainfall map and which are out of line with the deposition profile inferred by the other measured values. The points representing these three values have been left off Figure 5.1 in order to simplify the presentation of the results. The following discussion is concerned with sites C3, C5 and C9, the three anomalous sites.

Discussion of sites:

Site C3 at 1,050 m asl is located in a small depression a few metres in diameter, containing soil and a cover of grass, within a region of exposed bedrock. A high value of the ^{210}Pb(atmos) inventory was expected from this site due to possible collection of runoff from the surrounding rocks. The slope of the soil in the depression was small relative to the hill profile and there is a possibility that precipitation falling on the site passes over the surface, during heavy rainfall,

rather than percolating into the soil profile. In addition, the site may have been largely sheltered from the effect of occult deposition by the 'lip' of the rock bowl.

The results from site C5, a level piece of ground at 780 m asl, pose a problem in that the values of the ^{210}Pb(atmos) inventory from the two separate sampling points are different by about a factor of 3. Point (i) gives a value of $0.26 \pm 0.03\,\mathrm{Bq\,cm^{-2}}$ while point (ii) gives a value of $0.73 \pm 0.08\,\mathrm{Bq\,cm^{-2}}$. As the two sampling points were only a few metres apart this result suggests that different post-depositional processes have affected the ^{210}Pb(atmos) in the soil at the two points, or at least that very different rates of migration have been operating. Loss-on-ignition measurements, see Table 4.4, show that there is no shortage of organic matter to trap the ^{210}Pb(atmos) at sampling point (i). Although Urban et al. [149] have demonstrated that in the northeastern United States ^{210}Pb is mobilized by the organic-rich waters of peatlands, the ^{210}Pb is not lost evenly and most loss occurs primarily in hollows at depths below the water table. In such areas hummocks may give a good estimate of ^{210}Pb deposition from the atmosphere. It is possible that the results from site C5 may reveal a similar differential migration in waterlogged organic rich soils. The ^{210}Pb(atmos) inventory measured from point (ii) at this site is in line with the inferred deposition profile and will be included in the analysis.

Site C9 is clearly not retaining deposited ^{210}Pb(atmos). From the rainfall delivered to this site an inventory in the order of $0.34\,\mathrm{Bq\,cm^{-2}}$ is expected. The measured inventory was $0.013 \pm 0.007\,\mathrm{Bq\,cm^{-2}}$, which is only 4% of the expected value. Loss-on-ignition experiments showed that the soil at this site contained only a small proportion of organic matter by weight, see Table 4.4, and the capacity of this soil to trap deposited ^{210}Pb may be slight. The site was located on the flood plain within 100 m of the River Awe and may also have been prone to waterlogging and periodic flooding. Moreover, at this low altitude (50 m asl) there is also a possibility that the site may have been cultivated within the last few decades. These factors suggest, with hindsight, that site C9 was not a good choice for sampling. More generally, the selection of low altitude sites for ^{210}Pb(atmos)

inventory measurement has been a persistent problem since the soil must have been undisturbed for 50–100 years and in the UK such undisturbed sites are rare. It has been particularly difficult to find good low altitude sites at the locations in this study.

Deposition profile:

The deposition profile indicated by the measurements shows a steady increase with altitude. The highest value is seen to the east, ie downwind, of the mountain summit, a situation which is predicted by the model of Hill et al. [80] for the deposition of sulphate by seeder-feeder scavenging over 'short' hills, such as the five mountains of this study. This situation is in contrast to that at Great Dun Fell where the maximum ^{210}Pb(atmos) inventory is located at the summit. It must be noted that a smaller number of sites were sampled at Ben Cruachan than at Great Dun Fell, ie 9 as opposed to 23, and it is possible that had the transect at Ben Cruachan been extended further to the east then higher values for the ^{210}Pb(atmos) inventory would have been obtained. Although relatively few sites were sampled at Ben Cruachan, the results could indicate that occult deposition at the summit is not as important at this mountain as it seems to be for Great Dun Fell. An important difference between these two mountains, when considering occult deposition, is that Ben Cruachan is very rocky above $\sim$1,000 m asl. The summit region is mostly exposed bedrock with isolated patches of soil and grass. The summit region of Great Dun Fell has an almost unbroken cover of rough grass which has a much greater aerodynamic roughness than exposed rock. For the same mean windspeed, a greater degree of turbulence and thus a faster rate of occult deposition would be expected over the rougher, than the smoother, surface. At Ben Cruachan the displaced maximum of deposition suggests that seeder-feeder scavenging is dominant in producing the observed deposition profile.

Enhancement:

The lowest altitude site which gave a believable value for the ^{210}Pb(atmos) inventory was site C8 at 350 m asl. Mean annual rainfall at this site is shown to be 2,140 mm yr^{-1}. Mean annual rainfall at the western coast of Britain is

typically about 1,000 mm yr^{-1} so, at the lowest altitude site at Ben Cruachan, rainfall is already significantly enhanced. To calculate the enhancement of ^{210}Pb concentration in precipitation at the summit over that in the seeder rain and that in feeder cloud water over that in the seeder rain it is desirable to estimate the ^{210}Pb(atmos) inventory at the coast.

Sites C7 and C8 give a mean inventory of 0.37 ± 0.03 Bq cm^{-2} and a mean annual rainfall of 2,170 mm yr^{-1}. Assuming that the amount of ^{210}Pb deposited increases monotonically with rainfall amount, an inventory of $\sim 0.17 \pm 0.02$ Bq cm^{-2} would be expected with 1,000 mm yr^{-1} rainfall at the coast. This value of the inventory is in line with that found in the Eden Valley, at Great Dun Fell, and also with the inventory at Plymouth of 0.22 Bq cm^{-2} with 1,100 mm yr^{-1}, inferred from measurements of ^{210}Pb, in rainfall [35].

Combining the results from C1 and C2, the mean values of the ^{210}Pb(atmos) inventory and rainfall are 0.91 ± 0.14 Bq cm^{-2} and 3,480 mm yr^{-1} respectively. Comparing these values to those expected at the coast, the ^{210}Pb(atmos) inventory increases by a factor of ~ 5.4 and rainfall by a factor of ~ 3.5. Thus the concentration of ^{210}Pb in precipitation at the summit is greater by a factor of ~ 1.5 than the concentration in the seeder rainfall. Feeder cloud droplets would therefore contain a concentration of ^{210}Pb ~ 1.8 times greater than that in the seeder rainfall. Considering the occult deposition of cloud water at the summit, if an extra input of water amounting to 10% of the rainfall amount is assumed, which is probably an overestimate, then the concentration of ^{210}Pb in precipitation at the summit over the seeder rainfall becomes a factor of ~ 1.4 and the concentration of ^{210}Pb in cloud water over that in seeder rain becomes a factor of ~ 1.5.

It is interesting to calculate the concentration ratios for sites which are upwind of the mountain summit in order to make a comparison with the values obtained from the sites near the summit. Combining the values from site C4 and point (ii) at site C5, at 850 m and 780 m asl respectively, gives a mean ^{210}Pb(atmos) inventory of 0.80 ± 0.06 Bq cm^{-2} and a mean value of the annual rainfall of 2,500 mm yr^{-1}. These values indicate an increase in the ^{210}Pb(atmos) inventory of a factor of ~ 4.7

and an increase in rainfall amount of ~ 2.5. These figures show that the ^{210}Pb concentration in rainfall at these altitudes is a factor of ~ 1.9 greater than in seeder rain and the concentration in feeder cloud water is a factor of ~ 2.5 greater than in seeder rainfall. Assuming an occult deposition input of water amounting to 10% of that in rainfall, these values fall to factors of ~ 1.7 and ~ 2.1 respectively. Table 5.4 gives the intermediate values calculated for the summit and upwind of the summit ie the value between that for no extra input of precipitation and that for an extra input of water of 10% due to occult deposition.

Sites	feeder rain / seeder rain	feeder cloud / seeder rain
C1 + C2	~ 1.5	~ 1.7
C4 + C5(ii)	~ 1.8	~ 2.3

Table 5.4: The calculated ratio of ^{210}Pb concentration, feeder rain/seeder rain and the ratio of ^{210}Pb concentration, feeder cloud/seeder rain for sites C1 and C2 in combination and sites C4 and C5 (ii) in combination. The uncertainty associated with each value is ± 0.3

Table 5.4 shows that the ^{210}Pb concentration in precipitation falling at high altitude at Ben Cruachan is on average about a factor of 1.6 greater than that in the rain falling at low altitude and that the concentration of ^{210}Pb in cloud water is on average a factor of ~ 2 greater than that in the seeder rain. The ratio of the concentrations of ^{210}Pb in the cloud water to that in the seeder rain at the summit is ~ 1.7, and upwind of the summit ~ 2.3. Although the error bars associated with the measurements, ie ± 0.3, overlap, it appears possible that the ratio declines with altitude. This finding is consistent with an increase in size of the cloud droplets as they are carried up the side of the mountain, leading to a fall in the concentration of chemical species by dilution. In theory, the concentration of ^{210}Pb in cloud droplets should be a maximum near the cloud base and a minimum at the summit where the cloud droplets are largest. It is certainly the case that

the health of plants may be damaged by the occult deposition of cloud droplets containing large concentrations of acidic species and it appears that it is at the cloud base where the most concentrated, and thus most damaging, acidic droplets are present.

5.1.4 Beinn Dorain

At Beinn Dorain the average annual rainfall map shows only a marginal increase in the mean annual rainfall between the Auch Gleann to the southwest and the summit. The increase in precipitation is from 2,400 mm yr^{-1} in the valley to 2,800 mm yr^{-1} at the summit. As at Ben Cruachan the rainfall in the valley is enhanced above the $\sim$1,000 mm yr^{-1} expected at the western coast. This is not unexpected considering the mean height of the ground to the west of Beinn Dorain is $\sim$ 350 m and that the narrowness of the Auch Gleann (< 2 km) prevents westerly air from descending before encountering the Beinn Dorain mountain mass. The ^{210}Pb(atmos) inventory increases from 0.41 ± 0.04 Bq cm^{-2} at site D11 (210 m asl) to 0.73 ± 0.09 Bq cm^{-2} at site D1 (1074 m asl) a factor increase of $\sim$ 1.8.

Sites:

Sites D1–D4 are located along the spur which falls away to the north of the summit. Sites D5–D11 are located along a line which runs from east to west. The line of sites has the appearance of a right angle with one line running northwards from the summit to the corrie, 'Coire an Dothaidh', and the other from the corrie westwards to the Bridge of Orchy. Two of the sampling sites (D5 and D6) lie on a steep slope. At the high altitude sites the soils were of a predominantly mineral nature whereas at the low altitude sites the soils were very organic. At four of the sampling sites D1, D4, D7 and D11 three sets of cores were taken. The coefficient of variation ranged between 8–21% with a mean of 14%.

Deposition profile:

Although the measured values of the ^{210}Pb(atmos) inventory are clearly higher at sites D1 and D2 than those at sites D10 and D11, the values in between do not show a steady increase with altitude, see Figure 5.2. As with Great Dun Fell

the measurements give rise to at least two possible explanations of the observed deposition pattern.

The first explanation is illustrated in Figure 5.4a. The solid line represents the deposition profile inferred from the measured points. In this case a steady increase in the inventory with altitude is seen due predominantly to washout of low level cloud by rain falling from above, sites D3–D6 and site D10 are considered to give underestimates of the ^{210}Pb(atmos) inventory. It is possible that sites D3–D6 may underestimate the ^{210}Pb(atmos) inventory as a result of water running over the surface during heavy rains. In particular, sites D5 and D6 were located on a steep slope, of gradient $\sim$ 1:2, and overland flow may be significant at these sites. Also, site D10 does appear to give a low value, the rainfall here would be expected to support a ^{210}Pb(atmos) inventory akin to those of sites D11 and D9.

The second explanation of the observed deposition profile is shown in Figure 5.4b. In this case, washout of low level cloud leads to a smaller increase in deposition with altitude. At the summit, though, occult deposition of cloud droplets is important and enhances the ^{210}Pb(atmos) deposition significantly. As at Great Dun Fell, assuming that the surface roughness is uniform, the highest mean windspeeds and thus greatest occult deposition rates would be expected at the summit. Figure 5.4b shows the region of high occult deposition rates at the summit superimposed upon the seeder-feeder scavenging profile. If this interpretation is correct then the 'extra' occult deposition input to the summit region can be calculated. The individual sets of cores from sites D1 and D2 give a mean inventory of $0.76 \pm 0.07\,\mathrm{Bq\,cm^{-2}}$, sites D3 and D4 give a mean inventory of $0.55 \pm 0.04\,\mathrm{Bq\,cm^{-2}}$. Assuming that seeder-feeder scavenging, with some occult deposition, gives a similar inventory at the summit region then the 'extra' occult deposition input amounts to $\sim$28% of total deposition at these sites. It must be said, though, that this estimate of the importance of occult deposition is quite high in comparison to estimates of occult deposition, to moorland vegetation, made by other workers, eg. [45][59][77]. None of these workers have measured occult deposition rates at altitudes as high as the summit of Beinn Dorain (1,074 m

asl).

Although there is a clear increase in the ^{210}Pb(atmos) inventory between the valley and the summit, interpretation of the measured deposition pattern is not so clear. Fewer sites were sampled than at Great Dun Fell, (11 as opposed to 23), and the topography is more complex thereby making the interpretation less secure. The first possible explanation, see Figure 5.4a, depends on the assumption that 5 of the sites are underestimating ^{210}Pb deposition from the atmosphere. If the rainfall data are correct and the increase in rainfall amount between the valley and the summit is only a factor of ~ 1.2 then an increase in the ^{210}Pb(atmos) inventory due predominantly to seeder-feeder scavenging of ~ 1.8 seems rather large considering that some enhancement is already taking place in the valley. This interpretation may indicate that the rainfall increase beteween the valley and the summit is larger than is indicated on the 30 year average annual rainfall map [107]. The second interpretation, which considers seeder-feeder scavenging with a large input due to occult deposition superimposed on the summit region, provides a better fit to the measured points and also implies an increase in the deposition of ^{210}Pb(atmos) of ~ 1.5 due to seeder-feeder scavenging which is more in line with the rainfall increase shown on the rainfall map. The small number of sampling sites, however, make explanation of the observed deposition profile rather speculative.

Enhancement:

It is useful to calculate the enhancement of the ^{210}Pb concentration in precipitation and cloud water for comparison with data from the other sites. The four points sampled at sites D1 and D2 provide a mean inventory of $0.76 \pm 0.07\,\mathrm{Bq\,cm^{-2}}$, rainfall is $2{,}800\,\mathrm{mm\,yr^{-1}}$. Comparing these values with the estimates of the ^{210}Pb(atmos) inventory and rainfall at the coast, calculated to be $0.17\,\mathrm{Bq\,cm^{-2}}$ and $1{,}000\,\mathrm{mm\,yr^{-1}}$ respectively, shows that at the summit of Beinn Dorain rainfall is ~ 2.8 times greater and the ^{210}Pb(atmos) inventory is ~ 4.5 times greater than at the western coast. These values correspond to concentrations of ^{210}Pb in feeder cloud water being ~ 1.6 (± 0.3) times greater than in seeder rain and

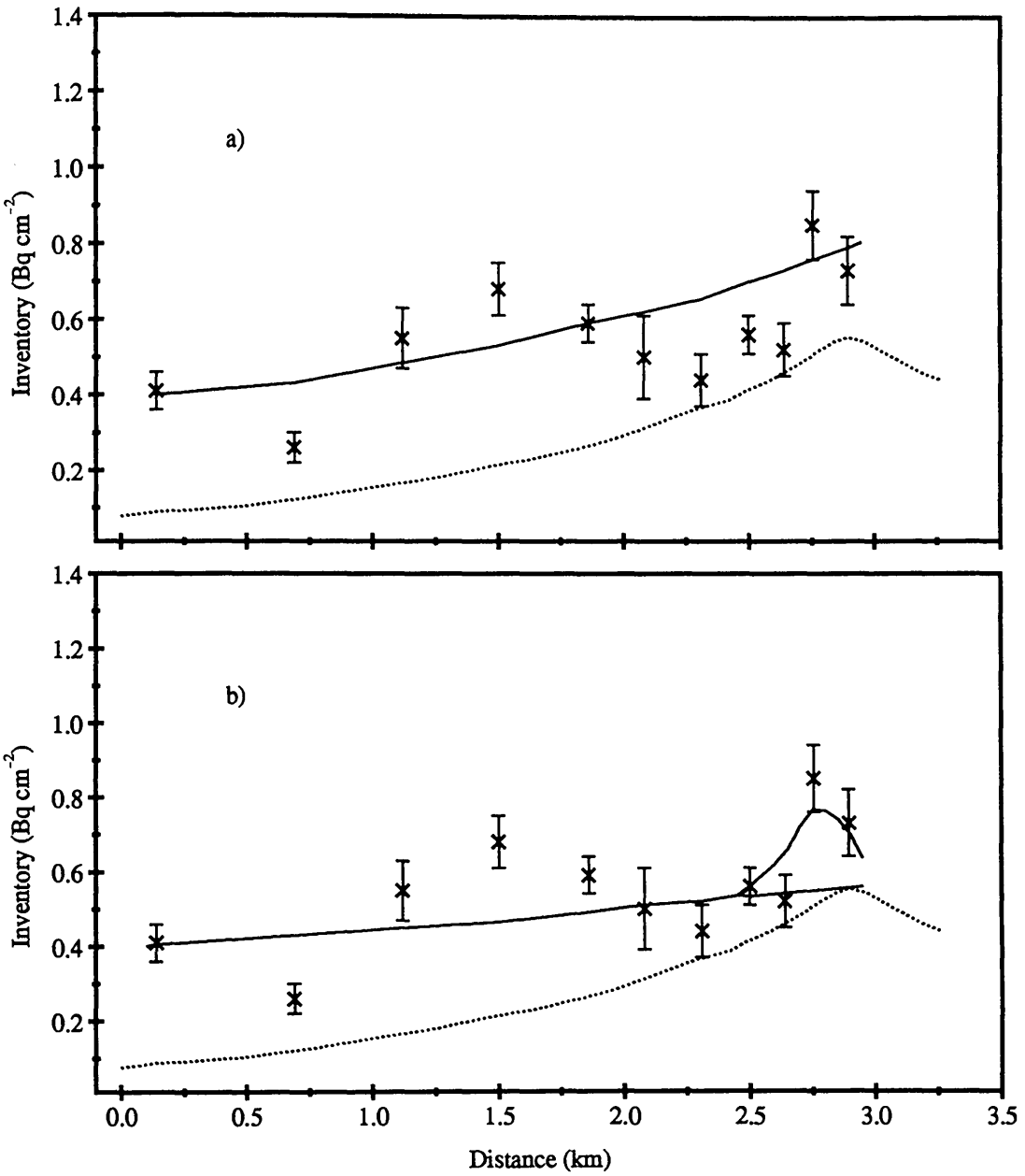

Figure 5.4: Two interpretations of the measured ^{210}Pb(atmos) profile at Beinn Dorain. The dotted line represents the profile of the hill and the solid line represents the deposition profile inferred by the measured points

the concentration in cloud water being ~ 1.9 (± 0.4) times greater than in seeder rain. Allowing for an input of water due to occult deposition amounting to 10% of annual rainfall, these values become ~ 1.5 (± 0.3) and ~ 1.7 (± 0.3) respectively. These values are similar to those calculated for Great Dun Fell and Ben Cruachan.

The lowest lying site, D11, which has an inventory of 0.41 Bq cm^{-2}, is subject to rainfall of 2,430 mm yr^{-1} which implies that rainfall amount has been enhanced by washout of cloud droplets. In other words, even in the valley, rainfall is enhanced by seeder-feeder scavenging. However, the ^{210}Pb(atmos) inventory measured does not point to the concentration of ^{210}Pb in rainfall being greater at this site than in the seeder rain. The ^{210}Pb(atmos) inventory observed at this site is that which would be expected with 2,430 mm yr^{-1} of rainfall which has the same concentration of ^{210}Pb as that falling at the coast. This suggests that the rainfall given for this site is an overestimate or that the ^{210}Pb(atmos) inventory measured is an underestimate. Alternatively, it could indicate that the value of 0.17 Bq cm^{-2}, calculated for 1,000 mm yr^{-1} of rainfall at the coast, is an overestimate.

5.1.5 Ben Lawers

Rainfall is shown, on the annual rainfall map, to increase from $\sim 1{,}490$ mm yr^{-1} at site L15 (200 m asl) to $\sim 2{,}500$ mm yr^{-1} at the summit. Thus, the rainfall at the summit of Ben Lawers is only about the same as that at the lowest sites at Beinn Dorain. These data represent a rainfall increase of a factor of ~ 1.7 between Loch Tay, to the south, and the summit. The line of the sites was northeast to southwest from the summit to site L12 and then in a north to south direction from site L12 to L15. Site L15 is situated 1 km from Loch Tay. However, the land to the west of the sampling line is very high. The mean height of the land between Ben Lawers and 20 km upwind, to the west, is ~ 680 m and this would be expected to have a profound effect on the deposition pattern if the dominant airflow, and most of the precipitation, originated from a westerly direction. Ben Lawers is situated within the Highland mass, roughly 80 km from the western coast, and, to some extent, is shielded from the western coast by mountains upwind. This location

cannot be treated as an isolated inland mountain and its rainfall and deposition pattern will reflect its complex topographical situation.

Sites:

At the five sites where 3 or more points were sampled, the coefficient of variation varied from 8–35% with a mean value of 18%. Site L11 gives a rather large value of the ^{210}Pb(atmos) inventory, and also for the ^{137}Cs(bomb) inventory.

Deposition profile:

The lowest four sites, L12–L15, give a very consistent value of the ^{210}Pb(atmos) inventory. Combining the results from these sites gives a mean inventory of $0.31 \pm 0.03\,\mathrm{Bq\,cm^{-2}}$ and rainfall of $1{,}650\,\mathrm{mm\,yr^{-1}}$. Above site L12 the deposition pattern appears highly variable. Sites L1, L7, L8 and L11 give relatively high values of the ^{210}Pb(atmos) inventory, ie $\sim 0.5\,\mathrm{Bq\,cm^{-2}}$, but sites L2–L6 give values which are more in line with the baseline value. The deposition pattern appears to be complex at the higher altitude sites.

Enhancements:

The four lowest sites show a small enhancement in rainfall concentration of ~ 1.1, and in cloud water concentration of ~ 1.4, over the concentration of ^{210}Pb in the seeder rain. Site L1, close to the summit, with a ^{210}Pb(atmos) inventory of $0.55 \sim 0.06\,\mathrm{Bq\,cm^{-2}}$ and rainfall of $2{,}500\,\mathrm{mm\,yr^{-1}}$, displays an enhancement in the concentration of ^{210}Pb in rainfall of ~ 1.3 and in cloud water of ~ 1.5. Assuming an extra input of precipitation amounting to 10% of the rainfall amount due to occult deposition of cloud droplets reduces these values to ~ 1.2 and ~ 1.3 respectively. Sites L7 and L8 combined give a mean ^{210}Pb(atmos) inventory of $0.48 \pm 0.04\,\mathrm{Bq\,cm^{-2}}$ and rainfall of $2{,}050\,\mathrm{mm\,yr^{-1}}$. These values correspond to an enhancement of ^{210}Pb concentration in rainfall of ~ 1.4 and in cloud water of ~ 1.7. Taking account of an extra 10% input of water due to occult deposition these enhancements become ~ 1.2 and ~ 1.4 respectively. On the other hand, sites L2–L6 do not show evidence of the deposition of ^{210}Pb being enhanced. The results from these sites may indicate a decrease with altitude of the concentration of ^{210}Pb in rain. The uncertainties associated with the concentration ratios given

above are of the order ± 0.3.

The variability at high altitudes indicates that the deposition pattern at Ben Lawers may be much more complicated than at Great Dun Fell, which is part of a ridge with a broad valley to the west, or the Merrick, which is a relatively isolated mountain. Local topographical factors may be affecting deposition at high altitudes, a high degree of variability is also seen for the deposition of ^{134}Cs at Ben Lawers. Ben Lawers is special in that it is higher than any of the other four mountains and the mean height of the land to the west is also relatively high, ie ~ 650 m asl. It may be that the deposition profile is almost flat but with some sites, ie L1,L7, and L8 attracting high rates of occult deposition due to local topographic features of the sites which enhance the turbulent deposition of cloud droplets. The solid line shown in Figure 5.2, which is considered to be a good fit to the measured points, indicates an inventory of $\sim 0.4\,Bq\,cm^{-2}$ at the summit. This would represent an increase of ~ 2.4 on top of the amount deposited with 1,000 mm yr^{-1} rainfall at the coast. The rainfall at the summit is indicated to be a factor of ~ 2.5 times greater at the summit than at the coast thus precluding an increase in the concentration of ^{210}Pb in rainfall with altitude.

The variability at high altitude makes inferring a deposition profile from the measurements difficult. The values, though, do seem to indicate very little or no enhancement in the concentration of ^{210}Pb in cloud and rainfall at Ben Lawers. Indeed, if a line which is almost flat were drawn through the measured points, then, with an increase in rainfall amount of ~ 1.7 between the valley and the summit, due to seeder-feeder scavenging during westerly airflows, a fall in the concentration of ^{210}Pb in cloud and rainfall with altitude would be indicated.

It has to be concluded that there is little evidence of seeder-feeder scavenging enhancing the deposition of ^{210}Pb with altitude at this mountain. This result was unexpected due to the height of Ben Lawers, (1,214 m asl), which was the highest mountain sampled, and the large vertical distance over which the samples were taken. Possible explanations of the observed ^{210}Pb(atmos) profile, at Ben Lawers, are discussed in the next section, within the context of the Highland transect.

5.1.6 Highland Transect

The three mountains Ben Cruachan, Beinn Dorain and Ben Lawers lie approximately on a line which runs from the western coast, near Oban, for about 80 km inland, see Figure 3.1. Ben Cruachan lies ~ 30 km from the coast and the line separating its summit from that of Beinn Dorain is about 26.5 km in length, on a bearing of 73°, which is roughly west southwest to east northeast. The line separating the summit of Beinn Dorain from that of Ben Lawers is about 31 km in length and lies on a bearing of 84°, roughly west–east. The relief along the line linking these mountains is shown in Figure 3.2. Although not directly on the coast there is relatively little high ground to the west of Ben Cruachan, in contrast to Ben Lawers where there is a good deal of high ground between it and the coast. The mean height of the land between the summit and 20 km to the west is 170 m for Ben Cruachan, 380 m for Beinn Dorain and 680 m for Ben Lawers (Table 5.1).

The mean annual rainfall amounts at each of the summits decrease with distance inland from the coast from 3,600 mm yr^{-1} at Ben Cruachan to 2,800 mm yr^{-1} at Beinn Dorain and 2,500 mm yr^{-1} at Ben Lawers. There is a corresponding decrease in the peak values of the ^{210}Pb(atmos) inventory from 1.05 Bq cm^{-2} at Ben Cruachan to 0.86 Bq cm^{-2} at Beinn Dorain and 0.55 Bq cm^{-2} at Ben Lawers. For rainfall amount the ratios of the values between the mountains, taking the value at Ben Cruachan as being unity, are; 1 : 0.78 : 0.69 and for the peak ^{210}Pb(atmos) inventory the relative values are 1 : 0.81 : 0.52. It seems that for Beinn Dorain the decrease in the peak, compared to the Ben Cruachan value, is similar to the decrease in the rainfall amount. However, it also seems that the decrease for the peak ^{210}Pb(atmos) inventory at Ben Lawers is greater than the decrease in yearly rainfall amount. This may indicate a 'cleansing' of the air upwind of Ben Lawers.

Taking the mean values of all the sites sampled at each mountain, the annual rainfall at Ben Cruachan is 2,730 mm yr^{-1} (the mean of the rainfall at sites C1–C8), 2,410 mm yr^{-1} at Beinn Dorain and 2,060 mm yr^{-1} at Ben Lawers. The mean of the ^{210}Pb(atmos) inventory is 0.65 ± 0.09 Bq cm^{-2} at Ben Cruachan, excepting

C5(i) and site C9, $0.55 \pm 0.05\,\mathrm{Bq\,cm^{-2}}$ at Beinn Dorain and $0.37 \pm 0.03\,\mathrm{Bq\,cm^{-2}}$ at Ben Lawers. Taking the values at Ben Cruachan as being unity, the relative sizes of the values are 1 : 0.88 : 0.75 for the yearly rainfall amount and 1 : 0.85 : 0.57 for the mean ^{210}Pb(atmos) inventory at each mountain. The pattern is similar to that with the peak ^{210}Pb(atmos) inventory and rainfall values. The mean ^{210}Pb(atmos) inventory at Beinn Dorain is reduced by the same amount as the yearly rainfall compared to the values at Ben Cruachan.

In contrast, the ^{210}Pb(atmos) inventory at Ben Lawers is a smaller fraction of that at Ben Cruachan than is the case for the mean yearly rainfall. Assuming that most of the rainfall is from the sector between west and southwest and that this rainfall is responsible for delivering the major fraction of the ^{210}Pb(atmos) present in the soil, then these results support the idea that feeder clouds are gradually depleted of liquid water by seeder-rain from above as they travel inland from the coast. In association with this 'drying' out of the feeder cloud there is also a 'cleansing' of the boundary layer air (ie a reduction in the concentration of the aerosol which carries ^{210}Pb and other chemical species, for instance SO_4^{2-}, NO_3^-, NH_4^+ etc). The results here suggest that by the time the Ben Lawers area is reached then the feeder clouds have been cleansed of ^{210}Pb bearing aerosols to a greater extent than they have been depleted of liquid water. Dore et al. [46] have observed the depletion of liquid water and cleansing of pollutant species from feeder clouds in experiments carried out at Corney Fell, a mountain near the coast in the Lake District, and Great Dun Fell, situated inland in the Pennines.

The ^{210}Pb(atmos) inventory between the valley and the summit increases at Ben Cruachan by a factor of ~ 2.8, by ~ 1.8 at Beinn Dorain and by < 1.7 at Ben Lawers. The value for Ben Lawers is calculated using the value of $0.55\,\mathrm{Bq\,cm^{-2}}$ from site L1. It thus appears that the factor increases between the valley and the summit decrease with distance inland. Comparing the values from each summit with the expected ^{210}Pb(atmos) inventory of $0.17\,\mathrm{Bq\,cm^{-2}}$ at the coast, gives a rough estimate that at Ben Cruachan the concentration of ^{210}Pb in rainfall is $\times 1.7$ that in rainfall at the coast, at Beinn Dorain the value is $\times 1.6$ and at Ben

Lawers it is less than $\times 1.3$. At Ben Lawers the value may even be < 1 if other sites except site L1 are considered.

These results indicate that similar enhancements of ^{210}Pb concentration in rainfall may be expected at Ben Cruachan and Beinn Dorain but that at Ben Lawers the enhancement due to seeder-feeder scavenging has all but petered out. In other words, the seeder-feeder scavenging effect is largely over by the time Ben Lawers is encountered. The feeder cloud is depleted of liquid water and ^{210}Pb by the time this mountain is encountered.

In addition to the depletion of ^{210}Pb and liquid water in the feeder-cloud as it is carried inland from the western coast, thereby reducing the enhancement of deposition on western slopes, rainfall originating from eastern sectors becomes more important towards the eastern coast. On the eastern coast of Scotland most of the rain which falls originates from eastern sectors [152]. At some point inland there is a 'watershed' which separates the region of Scotland where most of the precipitation is westerly in origin from the land where most of the precipitation is easterly in origin and it is believed that Ben Lawers may be situated close to this 'watershed' [133]. This would be expected to reduce the importance of seeder-feeder scavenging in enhancing deposition on the western slopes of Ben Lawers.

Figure 3.2 shows that to the west of Ben Cruachan and Beinn Dorain, the land is relatively low (~ 200 m altitude). Thus, air flowing from a westward direction, the direction from which most of the Atlantic frontal systems originate, will have to climb from a relatively low level to pass over the summit of the mountain and Ben Cruachan and Beinn Dorain stand out as individual mountains to the incoming frontal systems. In contrast, to the west of Ben Lawers is a great mass of high ground at > 600 m altitude and it appears possible that Ben Lawers does not stand out as an individual mountain but is shrouded by the high ground to the west. Pedgeley [123] discusses this effect for Snowdonia, in Wales, arguing that incoming frontal systems do not see a collection of individual mountains but instead see Snowdonia as a 'dome' with a few peaks rising above the dome. The effect of this

is to 'raise' the floor of the valleys, resulting in rainfall amounts in the valleys which do not reflect their true altitude. This effect arises because air flowing across a narrow valley will not descend into it but flow over the top. In valleys lying parallel to the airflow, funneling of air will occur. The measured inventory of ^{210}Pb(atmos) at Ben Lawers shows a pattern, ie little or no enhancement, which may reflect a similar situation as Pedgeley describes for Snowdonia. If this were the case then it would imply that the 30 year average rainfall map, which suggests a substantial increase in the amount of rainfall along the sampling line at this mountain, may be in error.

In conclusion, the measurements of the ^{210}Pb(atmos) inventory, at these 3 mountains, point to a decrease in the mean values with distance inland and also to a reduction in the increase with altitude of the ^{210}Pb(atmos) inventory with distance inland. These results indicate that the seeder-feeder scavenging mechanism declines in importance with distance inland from the western coast. Uncertainty in the rainfall field makes detailed interpretation of the deposition pattern speculative. However, the declining importance of seeder-feeder scavenging inland is thought to be a result of a combination of factors, specifically:

1. Drying out and cleansing of the feeder cloud as it is carried inland.

2. Frontal systems will respond to the Highland mountain mass as a whole rather than to individual mountains, as is possibly the case with Ben Lawers, ie the 'dome' effect.

3. At some point inland most of the rain falling originates from eastern sectors.

5.2 Relationship of ^{210}Pb(atmos) with Organic Matter

The weight loss on ignition was measured for all the samples taken from the Merrick and for the samples taken from sites C5 and C9 from Ben Cruachan. The

results are shown in Tables 4.1 to 4.4. The weight loss in each sample is assumed to correspond to the organic fraction (eg. [117]) and it is seen that the percentage of organic matter (by weight) decreases down the soil profiles.

For each of the samples subject to weight loss experiments the ^{210}Pb(atmos) per gram of dry soil (ie the organic and mineral fraction combined) has been calculated and in Figures 5.5, 5.6 and 5.7 these values have been plotted against the percentage of organic matter, for each sample. In Figures 5.5 to 5.7 the solid line represents the best straight line fit to the measured points. The details of the fits and the correlation coefficients are presented in Table 5.6. A correlation between an atmospheric input, for example of ^{210}Pb, and organic matter is often seen. It arises from the differences in soil density of organic (low) and mineral (high) soils. Thus, 1 g of organic soil represents deposit integrated over a larger area than 1 g of mineral soil [98].

At the Merrick each profile except those at sites M6 and M8 show a strong correlation between ^{210}Pb(atmos) g^{-1} and organic matter content. At site M8 a higher correlation coefficient would have been obtained if the lowest point had been ignored. Considering sites M1 to M6, where the sampling procedure was consistent, an interesting point is that the slope of the best fit line decreases with decreasing altitude. It is not known what process could produce this effect, if in fact the altitude dependence is a real effect. Nozaki et al. also observed a strong correlation between ^{210}Pb(atmos) g^{-1} and organic matter with each of 3 soil profiles analysed from beneath Pennsylvania woodland [117]. In addition, they discovered that the Pennsylvania profiles all followed the same trend, ie similar slopes, and that soils from Durban and New Mexico did not deviate markedly from this pattern. In contrast, the relationships at sites M1 to M6 display different slope values and point to a possible altitude dependence in the relationship.

Figure 5.7 shows the relationship between ^{210}Pb(atmos) and organic matter content for the Ben Cruachan sites C5 and C9. High values for the correlation coefficient are obtained from points (i) and (ii) at site C5 though the values for the slope are different, see Table 5.6. The samples analysed from site C9 show very

low values for the organic matter content and also very low values of ^{210}Pb(atmos) g^{-1}. It was not possible to fit a best straight line to the data from this site as the two points are too close together.

Atmospherically derived ^{210}Pb in soils becomes attached strongly to organic matter, see section in chap 2. For each of the sites at which loss on ignition experiments were conducted, the ^{210}Pb(atmos) per gram of organic matter was calculated, for each depth interval. The ^{210}Pb(atmos) g^{-1} of organic matter down the soil profile is given for site M6 and M8 in Table 5.5 The results from site M8 are typical in that the ^{210}Pb(atmos) g^{-1} of organic matter decreases with depth down the profile. This result contrasts with the finding of Lewis who found that the ratio was roughly constant, between the surface and 10–15 cm depth, at the forest sites he sampled in the northeastern USA [97]. Lewis attributed the constant ratio to rapid homogenization of ^{210}Pb(atmos) within the organic rich layers due, most likely, to physical mixing by animal organisms. Of all the sites analysed at the Merrick, and at Ben Cruachan, only at site M6 was the ^{210}Pb(atmos) g^{-1} of organic matter ratio roughly constant with depth, see Table 5.5. The results from the Merrick and Ben Cruachan indicate that rapid homogenization within the organic rich layers is not taking place at these sites. It is possible that animal activity within the soil is much less significant in the predominantly acid, moorland soils sampled in this study than in the soils studied by Lewis. These results certainly indicate that, in general, ^{210}Pb(atmos) is very strongly absorbed by organic matter at the top of the soil profile leaving little to migrate, and subsequently attach to organic matter, further down the soil profile.

Site M8		Site M6	
Depth (cm)	Bq g^{-1} of organic matter	Depth (cm)	Bq g^{-1} of organic matter
0–1	0.47	0–5	0.29
1–5	0.29	5–10	0.27
5–10	0.06	10–20	–
		20–25	0.30

Table 5.5: The concentration of ^{210}Pb(atmos) per gram of organic matter with depth at site M8 and site M6

Site	Number of points N	Correlation Coefficient r	Gradient m	Intercept c
M1	7	0.97	21.0 ± 1.7	-696 ± 72
M2	7	0.99	22.8 ± 1.9	-585 ± 64
M3	7	0.93	16.2 ± 1.3	-462 ± 60
M4	9	0.98	11.8 ± 1.0	-249 ± 31
M5	9	0.92	6.8 ± 1.2	-126 ± 33
M6	9	0.53	2.2 ± 1.4	8.9 ± 20.5
M7	5	1.00	7.9 ± 0.9	-69 ± 16
M8	5	0.58	2.2 ± 0.5	-119 ± 41
C5(i)	3	0.96	6.3 ± 0.7	-292 ± 44
C5(ii)	3	0.92	13.1 ± 0.8	-325 ± 30
C9	2	—	—	—

Table 5.6: Details of the best fit lines shown in Figures 5.5 to 5.7

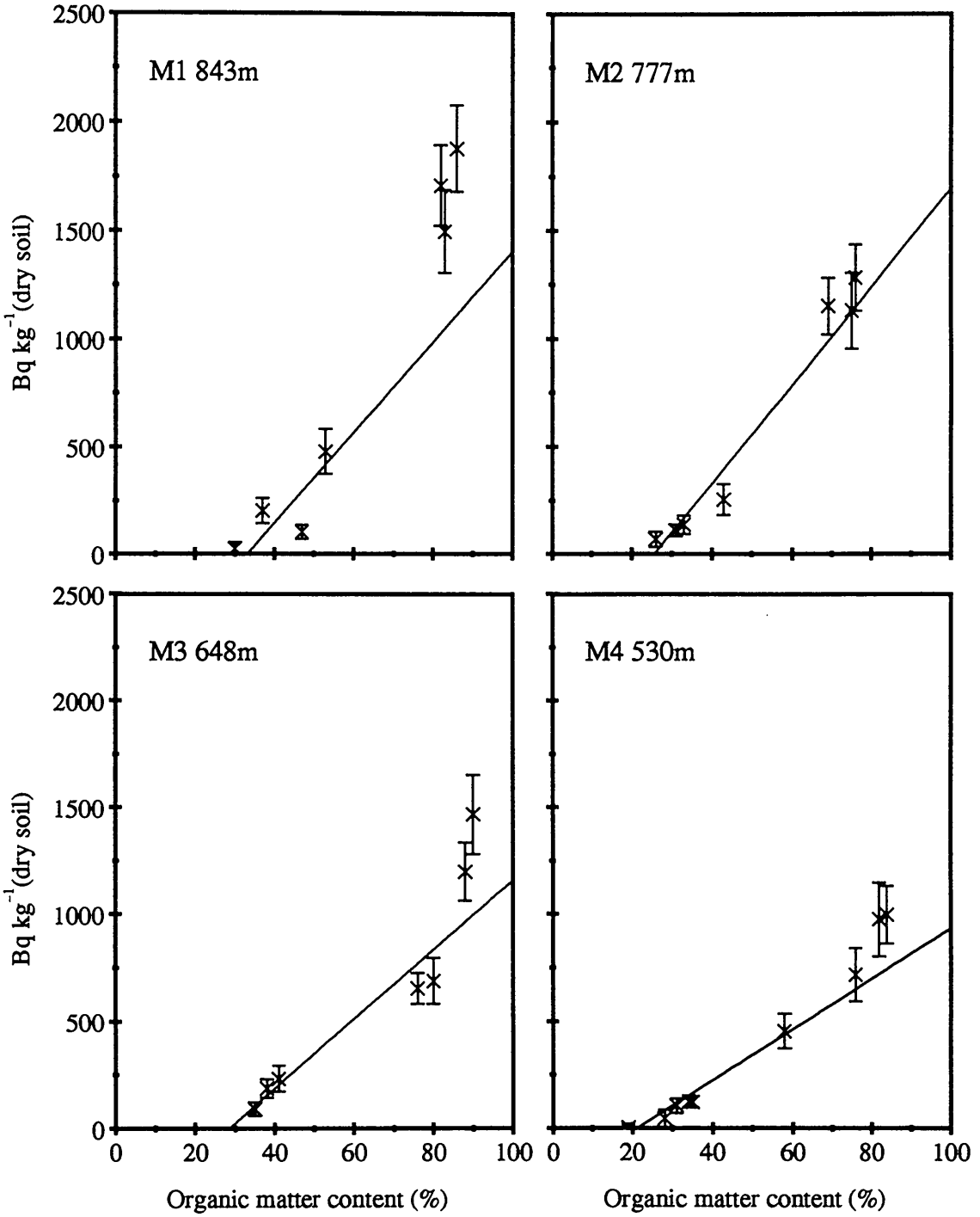

Figure 5.5: Relationship between ^{210}Pb(atmos) g^{-1} of dry soil and organic matter content for sites M1 to M4

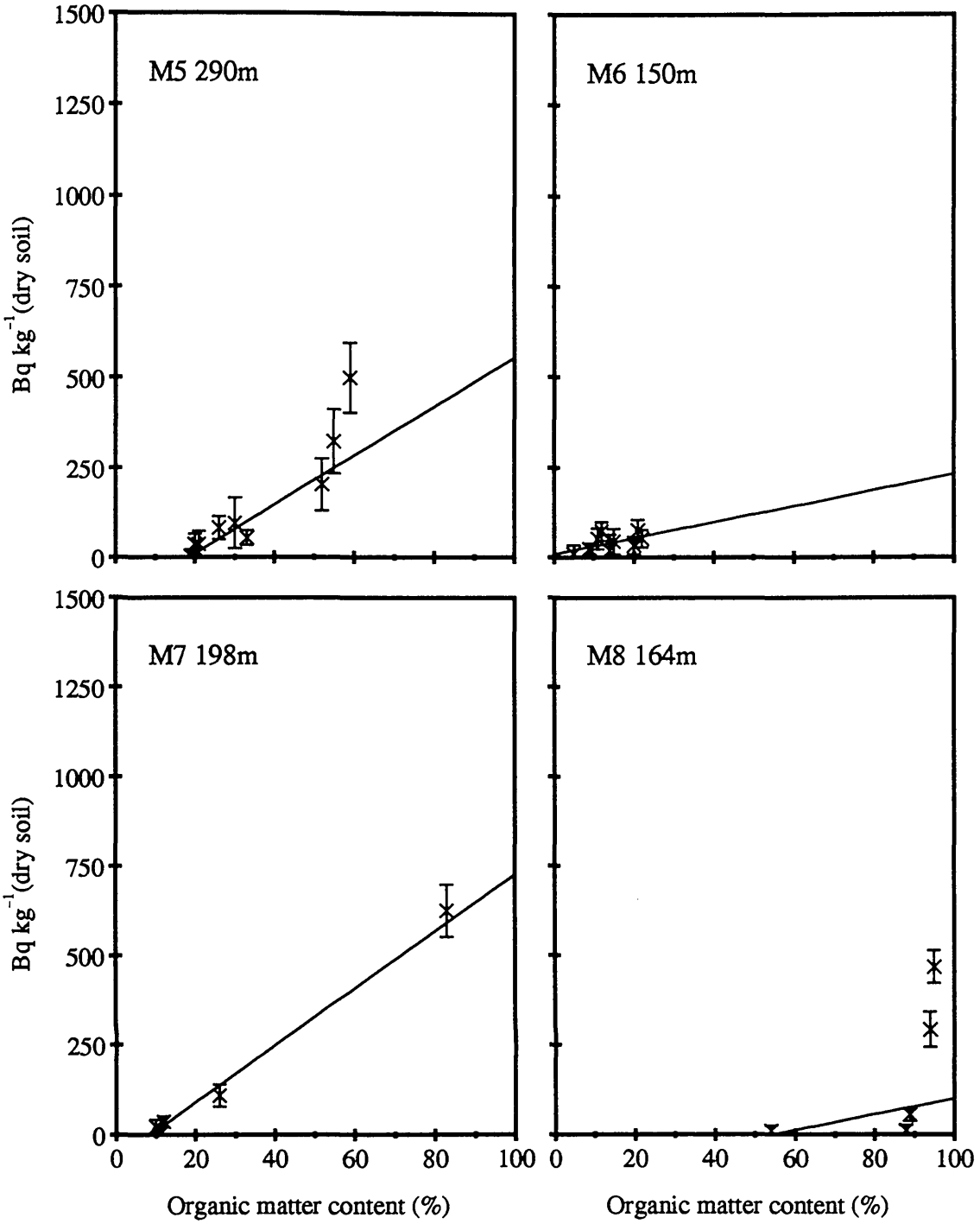

Figure 5.6: Relationship between ^{210}Pb(atmos) g^{-1} of dry soil and organic matter content for sites M5 to M8

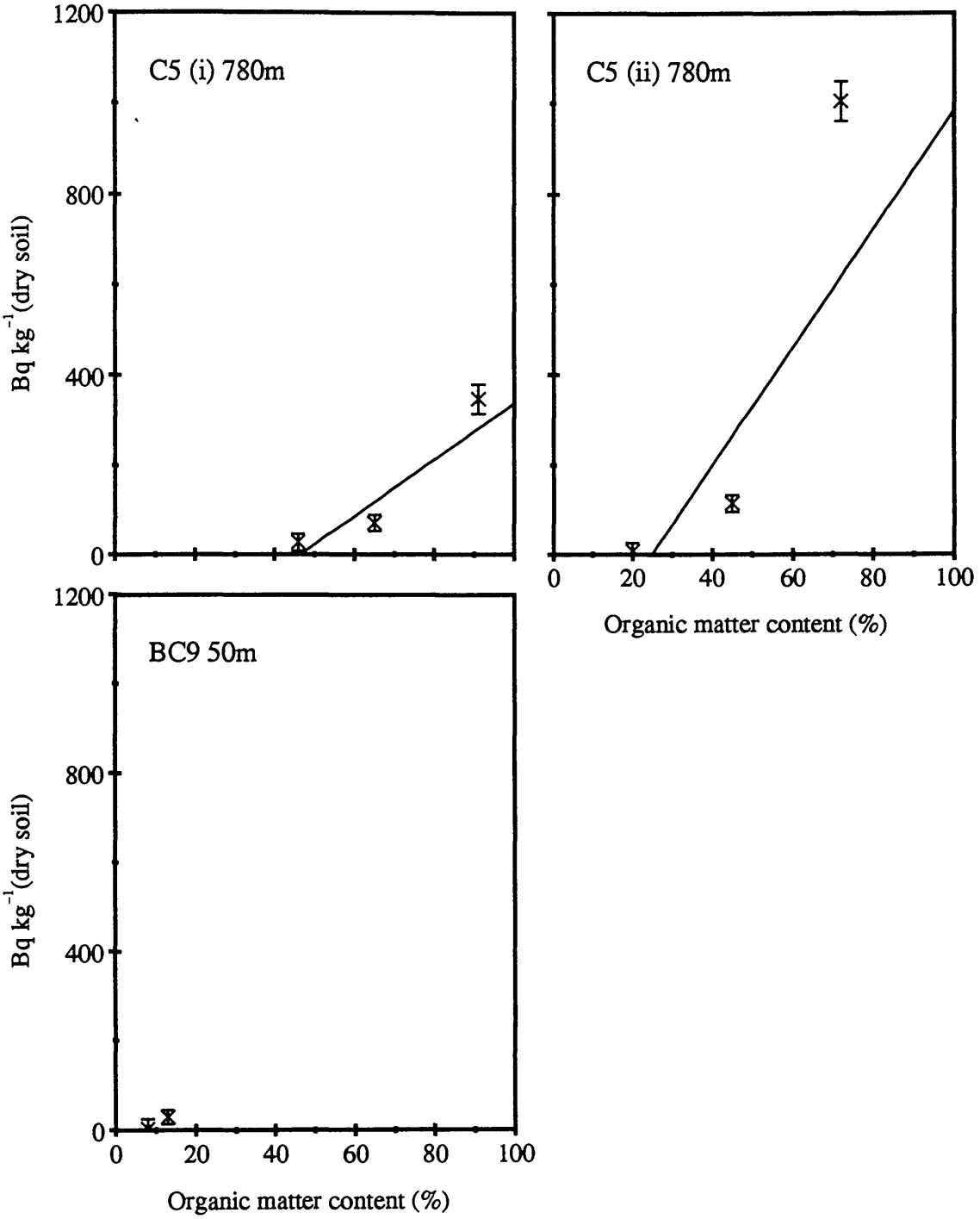

Figure 5.7: Relationship between ^{210}Pb(atmos) g^{-1} of dry soil and organic matter content for sites C5 and C9

5.3 Concentration of ^{210}Pb in Rainfall

By multiplying the measured inventory of ^{210}Pb(atmos) at each site by the decay constant, λ, an estimate of the total flux to the ground surface is obtained. By making the assumption that all the ^{210}Pb reaching the ground surface is delivered by raindrops it is possible to use the 30 year average annual rainfall map to gain an estimate of the average concentration of ^{210}Pb in rain at each of the sites. As a proportion of the ^{210}Pb(atmos) in the soil will have been delivered by the processes of dry and occult deposition, it is likely that this method will produce values of the average concentration which are overestimates. It must be remembered also that the data taken from the average annual rainfall map are uncertain by at least 10%.

Values of the average concentration of ^{210}Pb in rain have been calculated for low altitude sites and high altitude sites at each of the 5 mountains sampled in this study, see Table 5.7. In Table 5.7 the number in brackets represents the uncertainty in the measured ^{210}Pb(atmos) inventory. The actual uncertainty is larger due to uncertainties in the average annual rainfall data and in the proportion of ^{210}Pb(atmos) delivered to the soil by dry and occult deposition processes, which, for the purposes of this calculation , are assumed to be $< 10\%$. Underneath each value, in brackets, are written the sites from which the data were obtained. In Figure 5.8 the values of average ^{210}Pb concentration in rain at each site are shown for Ben Cruachan and Ben Lawers.

It is interesting in Table 5.7 that at Great Dun Fell, Ben Cruachan and Beinn Dorain the low altitude concentrations lie between 50–60 $\mathrm{mBq\,l^{-1}}$ and the high-altitude concentrations have values $\geq 80\,\mathrm{mBq\,l^{-1}}$. Thus, at these three mountains an increase in the concentration of ^{210}Pb in rain with altitude is indicated. This result supports the suggestion of seeder-feeder scavenging whereby rainfall at higher altitudes scavenges cloud, which frequently shrouds the upper slopes of the mountain, which contains relatively high concentrations of ^{210}Pb relative to concentrations in rainfall. The low altitude sites at Ben Lawers give a value

for ^{210}Pb concentration which is in line with those from the low-altitude sites at Great Dun Fell, Ben Cruachan and Beinn Dorain.

The values from the original Merrick sampling sites, M1–M6, are out of line with those from the other 4 mountains. The low altitude value of the average concentration of ^{210}Pb ($32 \pm 7\,\mathrm{mBq\,l^{-1}}$)is roughly half that of the other mountains and the high altitude value is only of the same order as the low altitude values from the other 4 mountains. The results from Merrick sites M1–M6 have been discussed earlier, see section 5.1.1. The later Merrick sampling sites M7 and M8 give a value of the concentration which is higher than that from sites M1 and M2 combined and which is also higher than the low altitude values from the other 4 mountains.

At Great Dun Fell the largest average ^{210}Pb concentrations are observed at the summit. However, as stated in section 5.1.2, it is likely that a large proportion of the ^{210}Pb(atmos) in the soil at the summit is there as a result of occult deposition, rather than deposition in rain. The results from Ben Cruachan are shown in Figure 5.8. At this mountain the largest concentrations of ^{210}Pb in rain are indicated to be at sites upwind of the summit. As stated earlier, cloud droplets will continue to grow as they are lifted up the mountainside, the concentration of ^{210}Pb in cloud droplets will be greatest near to cloud base and, due to dilution, the lowest concentrations in cloud are expected at the mountain summit. At Ben Lawers the concentration of ^{210}Pb in rainfall appears to be roughly constant with altitude. Indeed, if the results from sites L2–L6 are considered then a decrease in the concentration of ^{210}Pb in rain with altitude is indicated. It may be that the seeder-feeder mechanism is not an important process in the vicinity of Ben Lawers as this mountain is shielded from the western coast by roughly 80 km of the Highland mountain mass.

It is instructive to compare the values in Table 5.7 with those given in the literature for the UK. Pierson et al. [127] quote direct measurements of the ^{210}Pb concentration in rain at Milford Haven made during 1962 and 1963 of 80 and $87\,\mathrm{mBq\,l^{-1}}$. Eakins & Morrison [49] state that the average concentration of ^{210}Pb

Mountain	Low Altitude Sites Bq $l^{-1} \times 1{,}000$	High Altitude Sites Bq $l^{-1} \times 1{,}000$	Factor Increase
Merrick	32 ± 7 (M5,6)	57 ± 6 (M1,2)	1.8
	72 ± 9 (M7,8)		
Great Dun Fell	57 ± 12 (G6,7,9,10,11,16)	80 ± 6 (G19,20)	1.4
		102 ± 9 (G1,2,3,12)	1.8
Ben Cruachan	53 ± 4 (C7,8)	81 ± 2 (C1,2)	1.5
		99 ± 7 (C4,5ii)	1.9
Beinn Dorain	52 ± 6 (D11)	84 ± 8 (D1,2)	1.6
Benn Lawers	60 ± 4 (L12–15)	68 ± 10 (L1)	1.1
		43 ± 4 (L2–6)	0.7

Table 5.7: Average concentrations of ^{210}Pb in rainfall at the 5 mountains sampled in this study. The values are obtained from the measured inventory of ^{210}Pb(atmos) in the soil combined with data taken from the 30 year average annual rainfall map

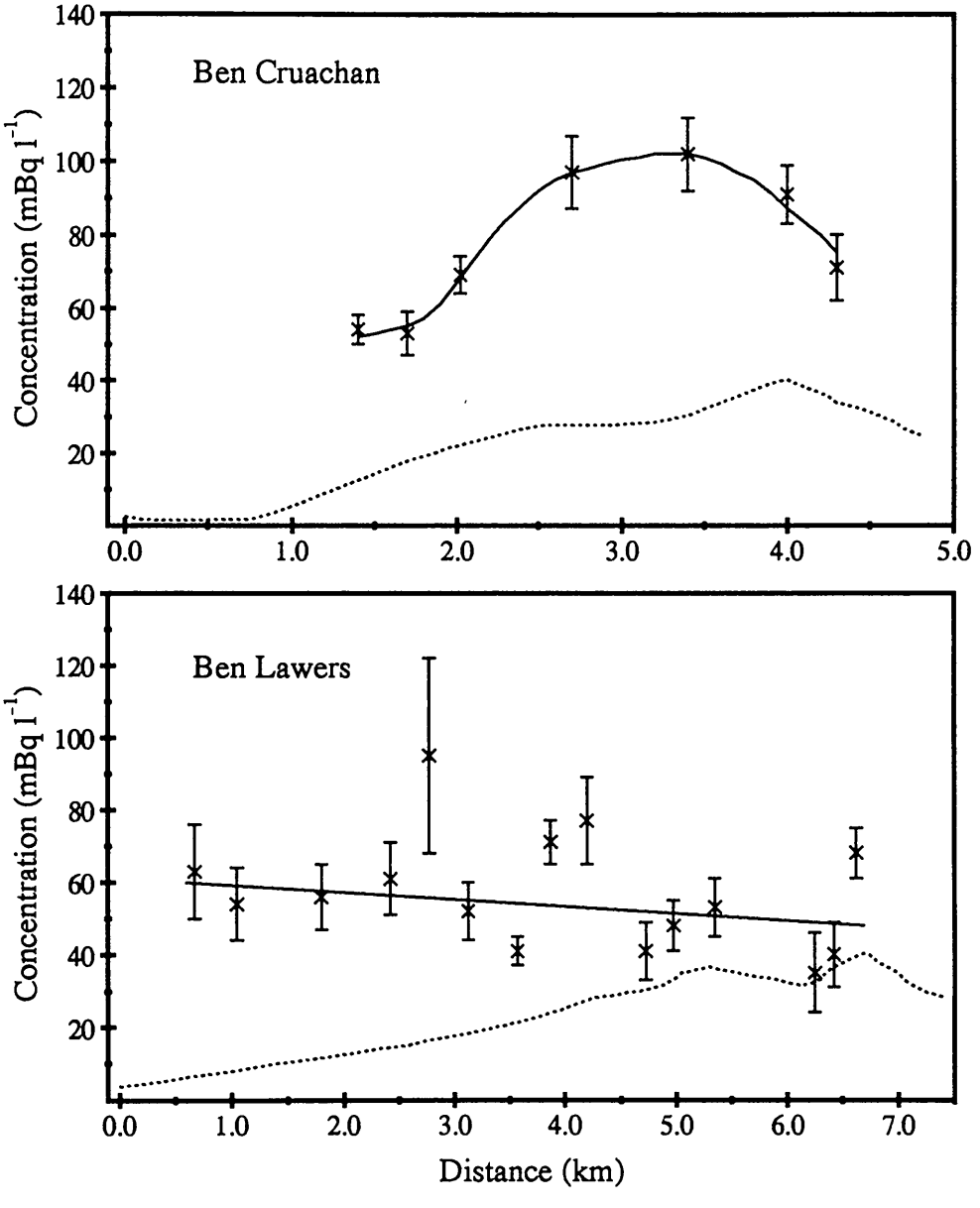

Figure 5.8: Average concentration of ^{210}Pb in rain (Bq l^{-1} × 1,000) at Ben Cruachan and Ben Lawers, calculated using the measured inventory of ^{210}Pb(atmos) in soil at each site and the Met. Office 30 year average annual rainfall map [107]

in rain, over the UK, is $\sim 74\,\mathrm{mBq\,l^{-1}}$. Clifton [35] measured the atmospheric ^{210}Pb flux at Plymouth between November 1988 and November 1990 and found it to be $6.8\,\mathrm{mBq\,cm^{-2}\,yr^{-1}}$. Using the value of $1{,}100\,\mathrm{mm\,yr^{-1}}$ for the average annual rainfall, as given on the 30 year rainfall map, produces a value of the average concentration of ^{210}Pb in rain of $61\,\mathrm{mBq\,l^{-1}}$. The values of ^{210}Pb concentration in rain for this study appear to be low compared to the values measured at Milford Haven but in line with those inferred from the flux measurements made at Plymouth.

Chapter 6

Discussion of the ^{137}Cs(bomb), ^{137}Cs(Chern) and ^{7}Be Results

In the preceding chapter the measured ^{210}Pb(atmos) inventories and annual inputs were discussed and interpreted to show how the mechanisms of wet deposition are modified by orography. These data have been used to validate assumptions made in estimating the deposition of acidic compounds to the uplands of the UK. During the course of this chapter the results for other radionuclides of interest will be discussed and interpreted. These include the fraction of ^{137}Cs which was delivered by the bomb testing program, ^{137}Cs(bomb), the fraction which originated in the Chernobyl accident, ^{137}Cs(Chern), and the cosmogenic nuclide ^{7}Be for which measurements made at the Merrick are discussed.

6.1 The measured inventory of ^{137}Cs(bomb) at each location

The total ^{137}Cs activity in each sample of soil, or vegetation, was separated, as described in section 4.1, into a fraction due to the atmospheric testing of nuclear weapons and a fraction due to the explosion which occured at the Chernobyl nuclear reactor plant on 26^{th} April 1986. The ^{137}Cs due to weapons testing was

injected predominantly into the upper troposphere and stratosphere during the 1950's and early 1960's, [126][131]. Fallout from individual weapons tests became well mixed throughout the northern hemisphere and was returned to the ground surface by multiple precipitation events over a number of years. For seven sites in the UK, situated far from any weapons test sites, Peirson & Salmon observed a linear relationship between the surface concentration of ^{137}Cs and mean annual rainfall [124]. In contrast to the weapons fallout, the ^{137}Cs due to the Chernobyl accident was injected into the atmosphere from ground level and was delivered to the UK ground surface mostly by precipitation events over 3–4 days during the first week of May 1986. Thus, the Chernobyl deposit of ^{137}Cs on the ground, unlike the ^{137}Cs(bomb), is not representative of average meteorological conditions. For these reasons, the ^{137}Cs(bomb) and ^{137}Cs(Chern) results are treated separately.

The range of values of the ^{137}Cs(bomb) inventory at each mountain are shown in Table 6.1. The highest and lowest values represent values obtained at an individual sampling point rather than values for a site as a whole. The ratio of the highest value to the lowest value is quite large at Great Dun Fell and Ben Cruachan. At each of these mountain locations, mean annual rainfall increases by no more than a factor of 2 between the valley and the summit. From the above discussion, it would be expected that the soil inventory of ^{137}Cs(bomb) will show the same factor increase as for the mean annual rainfall. Thus, the fact that the inventory of ^{137}Cs(bomb), at each mountain, shows a greater range of values than the estimated rainfall suggests that post-depositional processes are active in moving deposited ^{137}Cs laterally on the surface or within the soil or transporting it below the depth of sampling (typically 15 cm).

The coefficient of variation was calculated for each site at which three or more points were collected. Table 6.2 shows the range of values and the mean value for Great Dun Fell, Beinn Dorain and Ben Lawers. These values compare with those of Graustein & Turekian who found that the coefficient of variation of ^{137}Cs and ^{210}Pb(atmos) between separate cores at the same site is typically 15–20% [73].

By 1986, on average in Britain, the ^{137}Cs remaining from the weapons tests was

$\sim 3{,}200\,\mathrm{Bq\,m^{-2}}$ per 1,000 mm annual rainfall [13][23]. This value was obtained by sampling soil, down to 30 cm depth, in each of 29 $100 \times 100\,km$ grid squares [27]. This relationship is used to indicate whether the ^{137}Cs(bomb) inventory at each site reflects the atmospheric input or whether it reflects post-depositional movements of ^{137}Cs. Figures 6.1 and 6.2 show the measured inventories of ^{137}Cs(bomb) at each of the 5 mountains, on the same vertical scale. The solid line represents the ^{137}Cs(bomb) inventory estimated from the above relationship between ^{137}Cs(bomb) and mean annual rainfall.

In the following sections the ^{137}Cs(bomb) results at each mountain are discussed in turn.

6.1.1 Merrick

In Figures 4.1 and 4.2 it can be seen that for each of the original sampling sites, except site M3, the values of the ^{137}Cs(bomb) inventory from the core and the pit samples are essentially the same within the uncertainties. Figure 4.3 also shows the results from the later sampling sites M7 and M8. The value from site M8 is greater than that from site M6 and that from site M7 is greater than that from site M5. However, the values at these sites are not as far out of line with the values from the original six sites as is the case for ^{210}Pb(atmos). As was discussed earlier, the results from this mountain indicate that the ^{210}Pb(atmos) inventories at sites M1–M6 were systematically underestimated. In contrast, ^{137}Cs is much easier to measure than ^{210}Pb, by γ-ray spectroscopy, as it emits a γ-ray of relatively high energy (661.7 keV) with a high branching ratio (85.2%). Also measurement of this radionuclide is less sensitive to varying depth division down the length of the core. Looking at Figures 4.1 and 4.3, there is an apparent rise in the ^{137}Cs(bomb) inventory with altitude from sites M8 and M6. The core samples show a greater rise than do the pit samples. The core samples also point to a possible decrease in the ^{137}Cs(bomb) inventory at the highest two sites M1 and M2. The low inventories at sites M1 and M2 could be the result of overland flow during heavy rains or alternatively the physical removal down the profile of soil particles with

Mountain	^{137}Cs(bomb) Inventory ($Bq\,cm^{-2}$)		Factor Difference
	Smallest	Largest	
Merrick	0.10 ± 0.09	0.54 ± 0.18	5.4
Great Dun Fell	0.08 ± 0.01	1.25 ± 0.10	15
Ben Cruachan	0.08 ± 0.03	0.91 ± 0.06	12
Beinn Dorain	0.31 ± 0.04	1.23 ± 0.13	4
Ben Lawers	0.11 ± 0.04	0.73 ± 0.20	6.6

Table 6.1: The range in measured values of the ^{137}Cs(bomb) inventory at each mountain. Values represent individual points rather than sites

Mountain	Coefficient of variation	
	Range(%)	Mean(%)
Great Dun Fell	8–36	19
Beinn Dorain	8–43	23
Ben Lawers	9–22	14

Table 6.2: Range in values and mean of the coefficient of variation for the ^{137}Cs(bomb) inventory at Great Dun Fell, Beinn Dorain and Ben Lawers. The coefficient of variation was calculated for each site at which samples were collected from 3 or more points

^{137}Cs attached, termed translocation. The fact that there is no apparent defecit in the ^{210}Pb(atmos) inventory at these sites is observed indicates that the second process is the most likely cause of the apparent shortfall in ^{137}Cs(bomb).

See Figures 4.8 to 4.10 show the depth distribution of ^{137}Cs(bomb) with depth in the soil profile. From the pit samples it is clear that the major fraction of ^{137}Cs(bomb) resides within 10 cm of the ground surface, yet at sites M1,3,4,5 there is a fraction found below 10 cm. At site M6 the greatest bulk of the ^{137}Cs(bomb) is found in the 5–10 cm section. The core samples show the greatest concentration is in the 0–1 cm depth interval at sites M1,2,4,5. At sites M3 and M6 the greatest concentration is found in the 1–10 cm interval. It appears that at site M6 the ^{137}Cs(bomb) is 'smeared' down the profile, even though ^{137}Cs(bomb) was not measured below 10 cm at this site it is thought that significant migration down the profile has ocurred. The depth profiles from sites M1–M6 show that a greater proportion of ^{137}Cs(bomb) is found deeper in the soil than is the case for ^{210}Pb(atmos). Caesium-137 is more mobile in these upland soils than is atmospherically derived ^{210}Pb which is adsorbed strongly by organic matter.

It is instructive to look at the depth profiles for sites M7 and M8, see Figure 4.10. At site M7 the bulk of the ^{137}Cs(bomb) is found above 5 cm depth, at site M8 the distribution has been 'smeared' throughout a larger depth of the profile, and significant amounts are observed even in the 15–20 cm interval. In addition, twice the total inventory of ^{137}Cs(bomb) is found at site M7 than at M8. The rainfall amount delivered to these two sites is very similar and it can therefore be assumed that the amount of ^{137}Cs(bomb) delivered to these two sites was the same. From the total inventories and the depth profiles it appears that ^{137}Cs(bomb) is being transported down the profile at site M8. As has been stated previously, the soils at sites M7 and M8 are of quite diferent character, M8 being highly organic and marshy whilst M7 is more mineral in character and resting on better drained pasture land. Hilton et al. have shown that fibrous peats act as a 'slow release' source of ^{137}Cs, in the catchment of Devoke Water, Cumbria they found that streams draining fibrous peat sub-catchments contain more Cs than

those draining podzolic and ranker soils [81].

The values of the ^{137}Cs(bomb) inventory from the pit and core samples, at sites M1–M6, have been combined to give the mean values and are plotted in Figure 6.1 along with the values from sites M7 and M8. The solid line represents the inventory expected by applying the relationship of Cawse & Horrill [27] to the rainfall data. Most of the sites give measured values of the ^{137}Cs(bomb) inventory which are lower than the expected values. Within the uncertainties, on the measured inventory and on the expected inventory (ie the rainfall is uncertain by at least 10%), it appears that site M7 is retaining the input of ^{137}Cs(bomb) within the top 15 cm of the soil profile. The depth profiles of ^{137}Cs(bomb) at this site support this claim. The shortfall between the measured and expected values is greatest at the summit, at site M1, and in the valley, at sites M6 and M8. The lack of ^{137}Cs(bomb) in the top 20 cm of the soil profile at the summit may be due to leaching or tranlocation of soil particles or, alternatively, due to heavy rainfall flowing over the surface rather than percolating into the soil profile. However, leaching and translocation of ^{137}Cs are most likely as the ^{210}Pb(atmos) value at the summit is not reduced as would be expected if rainwater were not percolating into the soil. In contrast to site M8, which has already been discussed above, site M6 is very mineral in character and deficient in organic matter. The depth profile of ^{137}Cs(bomb) also shows that the atmospheric input is not being retained in the top 30 cm of the soil at this site. This site lies close to a river and waterlogging of the profile may be responsible for the fact that ^{137}Cs(bomb) is not retained here.

6.1.2 Great Dun Fell

Figure 6.1 shows the measured values of the ^{137}Cs(bomb) inventory for each site at Great Dun Fell, the solid line represents the inventory expected from the rainfall. The results at this mountain are very interesting. Basically, there appear to be two populations of sites at this mountain. Ten sites to the west of the summit give measured values of the inventory which reflect the expected inventory very closely. The remainder of the sites, especially sites G13 and G14 to the west and

sites G20, G22 and G23 to the east, give values which are significantly lower than those expected. Site G21 gives a higher value than expected, for the ^{137}Cs(bomb) inventory, and will be discussed later. The fact that some sites at this mountain retain ^{137}Cs(bomb) more strongly, in the top 15 cm of the soil profile, than other sites, may be related to the characteristics of the soil at the sampling sites. The soils at the ten sites, to the west of the summit, which reflect the atmospheric input of ^{137}Cs(bomb) are mineral in character, brown earth type, sandy loams and, in the valley, gleys. These mineral soils should contain sufficient illite to sorb ^{137}Cs. The soils at the sites which are not retaining ^{137}Cs very strongly, in the top 15 cm, are peaty soils. Fibrous peats contain effectively no illite, and even if sufficient illite is present to absorb Cs, organic matter interferes with the process, either by taking up Cs itself or by preventing the absorption of Cs by illite [101]. The results suggest that ^{137}Cs is being leached from the top 15–20 cm of these highly organic soils.

The sites to the west of the summit show an increase, in the ^{137}Cs(bomb) inventory, which is less than a factor of two between the valley and the summit. This is consistent with the vertical distribution of ^{137}Cs, which was injected, predominantly, into the stratosphere and upper troposphere and thus was not enriched in boundary layer air. During seeder-feeder scavenging events the feeder cloud was not enriched in ^{137}Cs with respect to the seeder cloud and as a result deposition was proportional to rainfall, as shown in Figure 6.1. In contrast the inventory of ^{210}Pb(atmos), which originates at the ground surface, increases by a greater factor, between the valley and the summit, than does rainfall.

The results from site G8 are quite interesting. At this site, situated beneath a small wood, the ^{210}Pb(atmos) inventory appears to be roughly twice that at neighbouring sites. However, the ^{137}Cs(bomb) inventory is of the same order as the inventory at neighbouring sites. This may be an indication of the capture, by the trees, of cloud droplets and large aerosols which contain high concentrations of ^{210}Pb relative to ^{137}Cs. The ratio of ^{210}Pb : ^{137}Cs in boundary layer air is expected to be greater than that for free-tropospheric air as the major source for ^{210}Pb is

the ground surface. Trees are very efficient scavengers of cloud droplets and large aerosols present in low level air in comparison to non-woodland vegetation.

6.1.3 Ben Cruachan

At Ben Cruachan, see Figure 6.1 there appears to be a large difference in the measured ^{137}Cs(bomb) inventory between the low altitude and high altitude sites. The factor difference appears to be similar to that for the ^{210}Pb(atmos) inventory. However, except at site C4 and sampling point (ii) at site C5, the measured values of the ^{137}Cs(bomb) inventory are significantly lower than expected. The depth profiles of ^{137}Cs(bomb) at sites, C6, C7, C8 and C9 are 'smeared' and indicate that significant downwards migration is taking place. It appears that the factor difference between the high altitude and low altitude sites is due to the lower sites retaining a smaller proportion of deposited ^{137}Cs(bomb), than the higher sites, in the top 15 cm of the soil profile. Interestingly, at site C5, points (i) and (ii) give very different values of the ^{137}Cs(bomb) inventory, as they also do for the ^{210}Pb(atmos) inventory. The ^{137}Cs(bomb) inventory at C5(ii) is in line with that expected from the rainfall at this site, a finding which supports the use of the ^{210}Pb(atmos) inventory, from this sampling point, in the previous discussion chapter. The small values of the ^{210}Pb(atmos) and ^{137}Cs(bomb) inventories measured at C5(i) may indicate that precipitation is flowing over the surface instead of percolating into the profile at this sampling point, alternatively erosion may be taking place, by wind or water, removing top soil and thus reducing the measured radionuclide inventories.

6.1.4 Beinn Dorain

The measured values of the ^{137}Cs(bomb) inventory at Beinn Dorain are shown in Figure 6.2. At first sight there appears to be an increase by a factor of < 2 in the inventory between the valley and the summit. However, sites D2–D4 give values which are not significantly greater than those at the low altitude sites. The soil

profiles indicate that ^{137}Cs(bomb) may be migrating downwards, below ~ 15 cm depth, at sites D2–D4 and at sites D8–D11. Most of the sites at this location give measured values of the ^{137}Cs(bomb) inventory which are lower than the expected values (solid line). Sites D1 and D7 give values of the inventory which are in line with those expected. However, the measurements from the individual sampling points at these two sites are quite variable. Sites D1 and D7 will be discussed later. Figure 6.1 may indicate that, as at Ben Cruachan, the low altitude sites are losing ^{137}Cs(bomb), from the top ~ 15 cm of the soil, at a faster rate than is the case for the sites at higher altitudes. The soils on the lower slopes of Beinn Dorain are quite peaty and it may be that they are less able to fix ^{137}Cs than the thinner, more mineral, soils on the higher slopes.

6.1.5 Ben Lawers

The measured values of the ^{137}Cs(bomb) inventory at Ben Lawers are shown in Figure 6.2. The large error bars associated with the values, at many of the sites, are due the large amounts of Chernobyl derived ^{137}Cs in the soil at this location. The points are very scattered and there is no evidence of a trend, in the levels of ^{137}Cs(bomb), between the base of the mountain and the summit. However, sites L15 to L12 show a possible decrease in the ^{137}Cs(bomb) inventory with altitude, between 200 m and 430 m asl. During sampling there was a noticeable change in the character of the soil and vegetation at these sites. Site L15 is pastureland with a brown earth type soil, as are sites L14 and L13, whereas L12 is a moorland site with very wet, fibrous peat. It is possible that the ability of soil to retain ^{137}Cs, in the top 20 cm of the profile, increases as the amount of organic matter decreases and/or the amount of illite (which sorbs ^{137}Cs irreversibly) increases. Again, most of the sites give measured values of the ^{137}Cs(bomb) inventory which are lower than those expected by applying the relationship of Cawse & Horrill [27] to the rainfall data. This is most likely due to migration of ^{137}Cs(bomb), by leaching or translocation of particles, beyond the lower depth of sampling. Site L11 gives a relatively high value of the ^{137}Cs(bomb) inventory and will be discussed later.

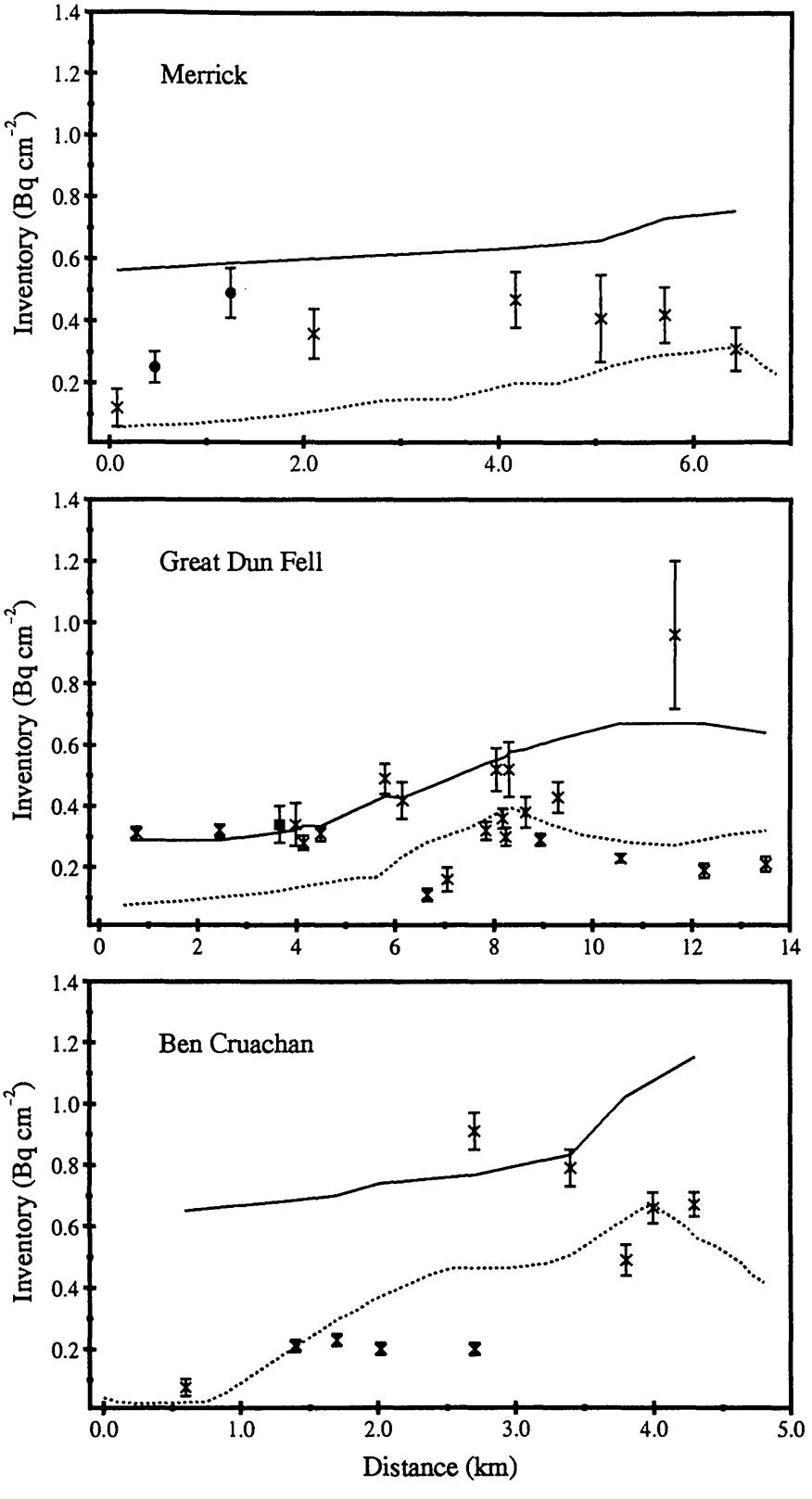

Figure 6.1: Inventory of ^{137}Cs(bomb) (Bq cm^{-2}) at the Merrick, Great Dun Fell and Ben Cruachan. The points represent the measured values and the solid lines represent the expected values

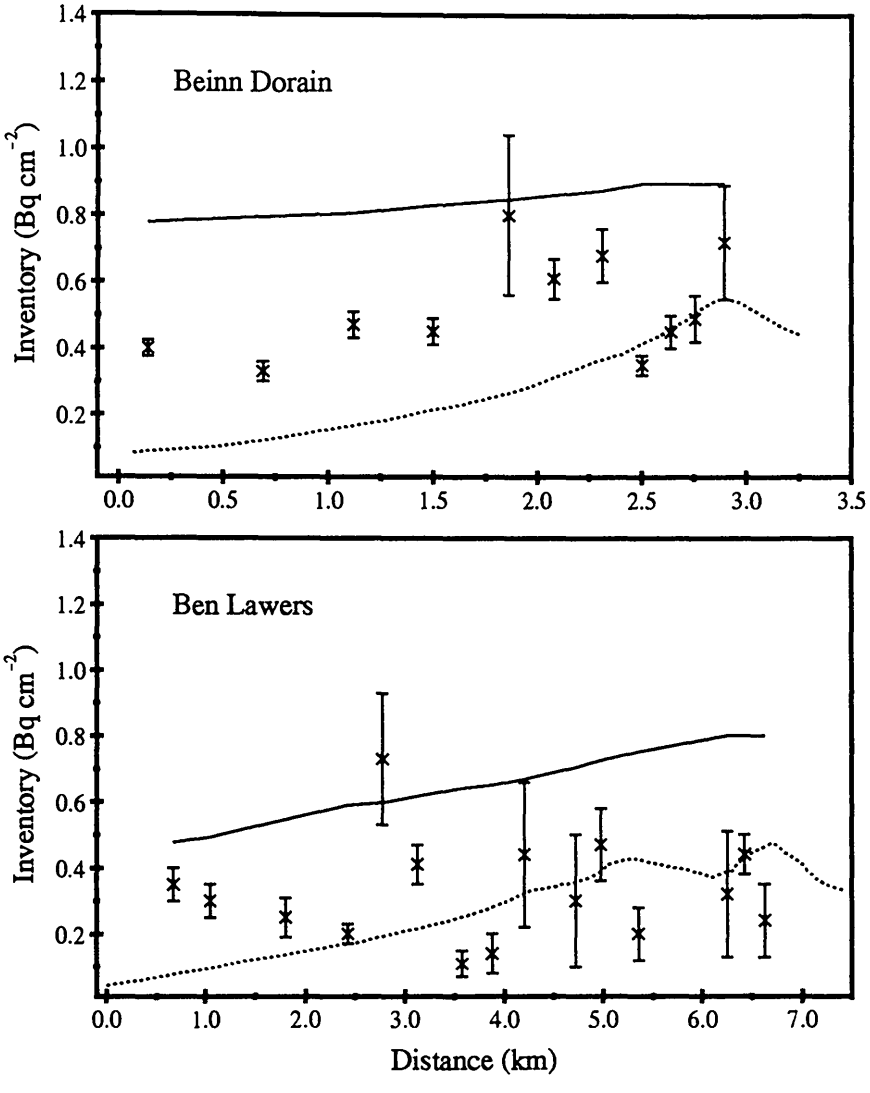

Figure 6.2: Inventory of ^{137}Cs(bomb) (Bq cm^{-2} at Beinn Dorain and Ben Lawers. The points represent the measured values and the solid lines represent the expected values

6.2 Comparison between expected and measured values of the ^{137}Cs(bomb) Inventory

At each mountain, the inventory of ^{137}Cs(bomb), measured in soil cores of average depth ~ 15 cm, has been compared with the inventory expected from the mean annual rainfall. On average, for the reference date of 1986, 3,200 Bq m^{-2} of ^{137}Cs(bomb) per 1,000 mm yr^{-1} rainfall is expected in the top 30 cm of the soil profile [23][27]. It is assumed, for the purposes of this study, that this figure is representative of the total atmospheric input of ^{137}Cs(bomb). This assumption is not unreasonable, for instance, Graustein & Turekian [73] took soil cores of 50 cm depth at 22 sites, in the northeastern USA, and found, on average, that 90% of the measured ^{137}Cs was present above 17 cm depth. This study is primarily concerned with finding sampling sites at which the measured radionuclide inventory reflects deposition from the atmosphere. Thus, we are concerned to find sites at which the bulk of the ^{137}Cs(bomb) inventory is being retained in the top 15–20 cm of the soil profile. Looking at Figure 6.1, a few sites appear to be retaining ^{137}Cs(bomb) within the top ~ 15 cm of the soil profile, for instance, site M7 and the ten sites at Great Dun Fell. However, as discussed in the previous section, most of the sites in this study do not give measured values of the ^{137}Cs(bomb) inventory representative of atmospheric deposition.

Table 6.3 shows the amount of ^{137}Cs(bomb) measured as a percentage of that expected from the average annual rainfall. At Great Dun Fell if only the 12 sites which give values smaller than expected are considered then the level of ^{137}Cs(bomb) observed compared to that expected becomes 47%. It can be seen that only about half, with the exception of Great Dun Fell, of the ^{137}Cs(bomb) expected down to 30 cm depth in 1986 was measured at each mountain. A similar finding was published by Bonnett & Appleby for the upland Llygad Rheidol catchment in mid-Wales [13]. They measured only 63% of the ^{137}Cs(bomb) expected to be present in the soil based on the mean annual rainfall. These results probably reflect the enhanced mobility of ^{137}Cs in the highly organic, peaty soils charac-

Mountain	Measured / Predicted ^{137}Cs(bomb) (%)
Merrick	55
Great Dun Fell	75
Ben Cruachan	51
Beinn Dorain	61
Ben Lawers	51

Table 6.3: The level of ^{137}Cs(bomb) measured at each mountain as a percentage of that predicted from the average annual rainfall using the relationship of Cawse & Horrill [27]

teristic of many of the sites sampled in this study. For soil samples of 15–20 cm depth, in the upland areas of this study, the relationship of Cawse & Horrill [27] overestimates the level of ^{137}Cs(bomb) present by about a factor of 2.

6.2.1 Outstanding values

Four sampling sites which attract particular interest are sites, G21, D1, D7 and L11 as at these sites levels of ^{137}Cs(bomb) have been measured which appear to be out of line with neighbouring sites at the same mountain. The values of the ^{137}Cs(bomb) inventory at each of these sites are shown in Table 6.4. In the case of sites G21 and D7 values are obtained which are also higher than would be expected from the annual rainfall, see Table 6.5.

The sampling points which show unexpectedly large inputs are points (i) and (ii) at site G21, point (ii) at site D1 and point(i) at site D7. At these locations the ^{210}Pb(atmos) inventories are not exceptionally high compared to values at neighbouring sites. A single point was sampled at site L11, but here the ^{210}Pb(atmos) inventory is also quite high in comparison to adjacent sites. This result indi-

Sampling site	^{137}Cs(bomb) Inventory at each point (Bq cm^{-2})		
	(i)	(ii)	(iii)
G21	1.14±0.09	1.25±0.10	0.49±0.04
D1	0.53±0.05	1.05±0.07	0.57±0.05
D7	1.23±0.13	0.39±0.05	0.78±0.08
L11	0.73±0.20		

Table 6.4: The values of the ^{137}Cs(bomb) inventory at the sampling sites G21, D1, D7 and L11

cates that this site is possibly receiving an extra input of precipitation water due, possibly, to overland flow off the surrounding terrain.

Table 6.5 shows the ratio, of the measured ^{137}Cs(bomb) inventory over that expected from the rainfall, for each of the sampling points. It can be seen that, at some of the sampling points, the values measured are substantially larger than would be expected from the rainfall data, eg points (i) and (ii) at site G21 and point(i) at site D7.

As discussed earlier, at many of the sites in this study less ^{137}Cs(bomb) than expected has been measured. For example at site M6 only 21% of the ^{137}Cs(bomb) inventory expected was observed, at sites G13 and G14 the amounts were 33% and 24% respectively, at site C9 the amount was 12%, at site D4 39% and at site L9 only 16% of that expected was observed. At a few sites, see Table 6.5, higher levels of ^{137}Cs(bomb) have been measured than are expected from atmospheric deposition alone. Atmospherically deposited ^{137}Cs may be carried away in solution by overland flow before getting a chance to sorb to organic colloids or fine particles within the soil. If attachment within the soil is not very strong, ^{137}Cs may be gradually lost in solution via groundwater flow, ie the soil acts as a 'slow release' source for ^{137}Cs [100]. Alternatively, ^{137}Cs attached to fine particles and organic

Sampling site	^{137}Cs(bomb) meas/exp		
	(i)	(ii)	(iii)
G21	1.8	2.0	0.8
D1	0.6	1.2	0.6
D7	1.5	0.5	0.9
L11	1.2		

Table 6.5: The ratio of the ^{137}Cs(bomb) inventory, measured/expected, for each of the sampling points at sites G21, D1, D7 and L11

colloids may be eroded from the soil surface or undergo translocation within the soil profile [11]. Thus, ^{137}Cs on the ground and in the soil may be redistributed through various post-depositional processes.

In the acid organic soils, which characterise many of the sites in this study, ^{137}Cs is known to be highly mobile relative to other soil types. It is possible that the sampling points where larger than expected values of ^{137}Cs(bomb) were observed are zones of redeposition of fine particles eroded from slopes higher up, or they may be a focus for overland flow carrying ^{137}Cs in solution. Site G21 was situated on a bluff with a gentle slope down towards the stream. Site D7 was situated near the foot of an extensive steep slope. Both these sites may be expected to experience overland flows and possibly deposition of radionuclides in solution or redeposition of fine particles eroded from higher slopes. Bonnett et al. noted higher than expected levels of ^{137}Cs at certain sites in the Wye catchment which they attributed to redeposition of fine particulate overbank sediments [12].

In summary, the ^{137}Cs(bomb) inventory at most of the sites in this study is not representative of the total atmospheric deposition. In contrast to ^{210}Pb, which is retained very strongly by organic matter, and is present, predominantly, in the top 15 cm of the soil profile, ^{137}Cs is very mobile in the highly organic soils of

the uplands of northern Britain. The evidence suggests that a significant fraction of ^{137}Cs(bomb) has been transported, by leaching or translocation processes, to depths greater than $\sim$15 cm at many sites. In order to use soil as a collector of the total deposition of ^{137}Cs(bomb) from the atmosphere, attention must be paid to its character, for instance fibrous peat should be avoided if possible, it being low in the minerals which sorb ^{137}Cs. However, even in soils which are not peat, strong ^{137}Cs retention is not assured; there is a complex interplay between Cs^+, K^+, H^+, clays and organic matter which may or may not combine to lock up Cs and prevent leaching [98]. In sampling, care must be taken to go deep enough to collect the bulk of the ^{137}Cs(bomb) inventory. Sampling down to bedrock would be ideal, but is impractical in many situations.

6.3 Contrasting the inventories of ^{137}Cs(bomb) and ^{210}Pb(atmos)

The source of the ^{137}Cs(bomb) present in the atmosphere over the UK has been, predominantly, the upper troposphere and stratosphere. In contrast, the ^{210}Pb in the atmosphere originates from the ground surface via ^{222}Rn emanation. Assuming that the concentration of ^{137}Cs(bomb) increases with altitude, or is roughly constant, within the troposphere and that the concentration of ^{210}Pb decreases with altitude (ie the ratio of ^{137}Cs(bomb)/^{210}Pb(atmos) increases with altitude in the troposphere) then it is possible that the ratio of ^{137}Cs(bomb)/^{210}Pb(atmos) in soil may give information on the vertical scavenging pattern of these radionuclides from the atmosphere [73][111]. Any change in the ^{137}Cs(bomb)/^{210}Pb(atmos) ratio in soil, with altitude, may indicate a change in the atmospheric scavenging pattern over high ground compared to low ground.

The preceding results show that ^{137}Cs is quite unlike ^{210}Pb and that most of the sites do not reflect the deposition of ^{137}Cs(bomb), due, most likely, to post-depositional migration within the profile. However, there are 10 sites on

the western slopes of Great Dun Fell which hold ^{137}Cs(bomb) inventories in line with those expected from mean rainfall. The ^{137}Cs(bomb)/^{210}Pb(atmos) ratios for these sites, excepting site G8, have been calculated and plotted against altitude in Figure 6.3. The solid line represents the best fit straight line to the measured points. The ratio shows a decrease with altitude from just under 2.0 in the Eden Valley to ~ 1.0 at the mountain summit. The correlation coefficient has a value $r = -0.61$.

Graustein & Turekian examined the ^{137}Cs(bomb)/^{210}Pb(atmos) ratio in forest soils in the northeastern USA [73]. They found the ratio to decrease from a mean value of 0.81 in lowland soils to a mean value of 0.66 at sites $> 1{,}000$ m altitude. Monaghan sampled Californian soils and found that the ^{137}Cs(bomb)/^{210}Pb(atmos) ratio ranged from 1.9 at the coast to 0.9 in the Sierra Nevada mountains, $> 2{,}500$ m altitude [110]. These results indicate that the increase in the level of ^{210}Pb(atmos) in soil with altitude is greater than for ^{137}Cs(bomb). Since the ratio in air of ^{137}Cs(bomb)/^{210}Pb increases with altitude, in the lower part of the troposphere, an explanation of this observation is that, scavenging of boundary layer air is more effective over higher ground than over lowlands. Seeder-feeder scavenging and occult deposition could both account for the increased scavenging of boundary layer air over high ground.

It is interesting that the range of values of the ^{137}Cs(bomb)/^{210}Pb(atmos) ratio at Great Dun Fell is similar to the range of values found in California by Monaghan, rather than those found by Graustein & Turekian for the northeastern USA. This could reflect the fact that both the UK and California are both situated on the western edge of continental land masses, and that scavenging processes related to precipitation and cloud deposition are similar.

In Figure 6.4 the inventory of ^{210}Pb(atmos) for each site at Great Dun Fell has been plotted against the inventory of ^{137}Cs(bomb). It can be seen that there is no clear pattern. The correlation coefficient has a value $r = 0.04$, indicating no relationship between the inventories of both nuclides. However, plotting the values from the 9 grassland sites to the west of the summit which are apparently retaining

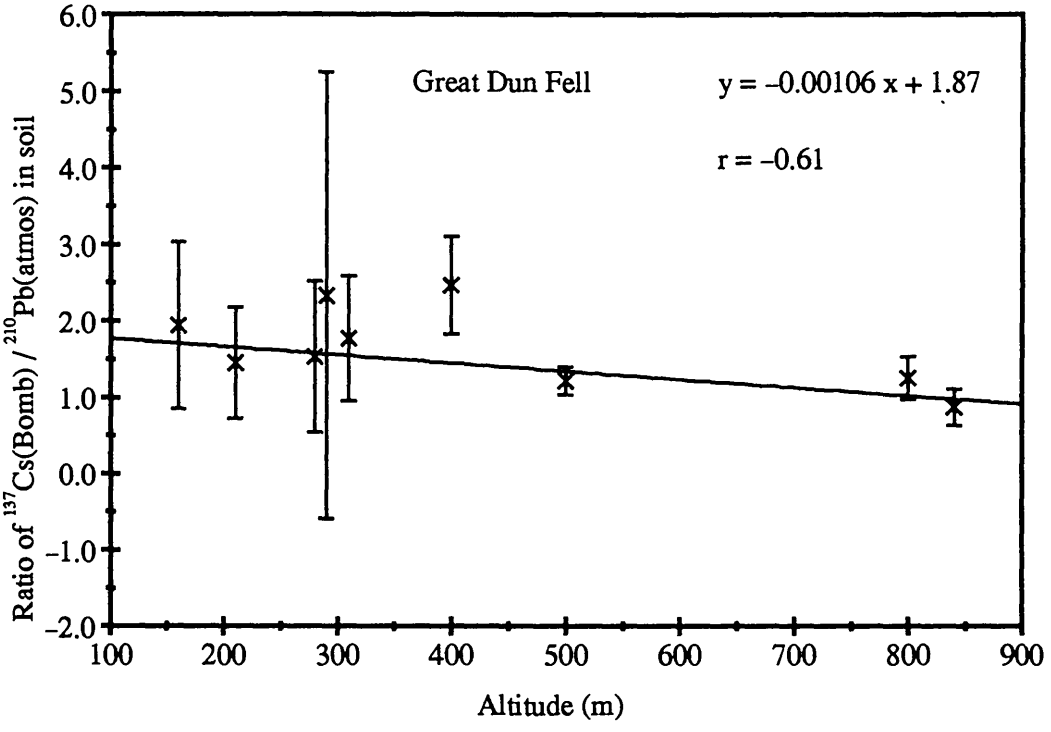

Figure 6.3: Plot of the ^{137}Cs(Bomb) / ^{210}Pb(atmos) ratio in soil versus altitude for 9 sites on the western slopes of Great Dun Fell. The sites represented are G1,4,6,7,9,10,11,15,16. These sites reflect the atmospheric deposition of ^{137}Cs(bomb)

^{137}Cs(bomb) gives a much clearer picture, see Fig 6.5. The correlation coefficient now has a value r = 0.82, indicating a significant positive relationship between the ^{137}Cs(bomb) and ^{210}Pb(atmos) inventories on the western slopes of Great Dun Fell. The small values represent the low altitude sites and the large values represent the high altitude sites. As the higher altitude sites are expected to receive a greater proportion of aerosols which have been scavenged from boundary layer air, by occult deposition and seeder-feeder scavenging, than the low level sites, it is expected that the ^{210}Pb(atmos) inventory should increase at a greater rate than the ^{137}Cs(bomb) inventory. Admittedly, the high degree of scatter permits the imposition of a variety of different lines on the measured points. However, a best fit line has been calculated (see solid line) and a line representing the two soil inventories increasing at the same rate has been drawn on the plot (see dotted line). The best fit line, on Figure 6.5, gives a slope quite different from the dotted line (representing the same rate of increase), and shows the ^{210}Pb(atmos) inventory to be increasing at a rate $\sim$1.8 times greater than the ^{137}Cs(bomb) inventory.

This result is further evidence for the enhanced scavenging of boundary layer air by Great Dun Fell, in comparison to adjacent lowlands. Migration of bomb derived ^{137}Cs below the sampling depth, at most of the sites sampled in this study prevents a comparison of the ^{137}Cs(bomb) and ^{210}Pb(atmos) inventories for the other mountains of this study.

6.4 Chernobyl Deposition

The radioactive plume from Chernobyl crossed Britain from southeast to northwest during 2–3 May 1986. It travelled out into the Atlantic and through recirculation crossed Britain again from southwest to northeast during 7–8 May [140]. Deposition of radioactive caesium was very patchy over the UK and Clark & Smith showed that levels of deposited ^{137}Cs can be closely related to rainfall intercepting the plume [33]. Deposition of ^{137}Cs was greatest in the north and west of the UK.

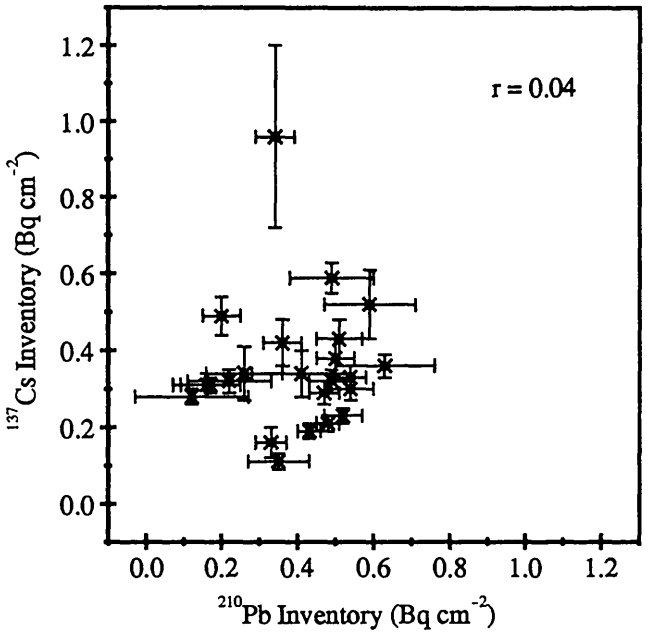

Figure 6.4: Plot of the ^{137}Cs(Bomb) inventory against the ^{210}Pb(atmos) inventory for all 23 sites at Great Dun Fell

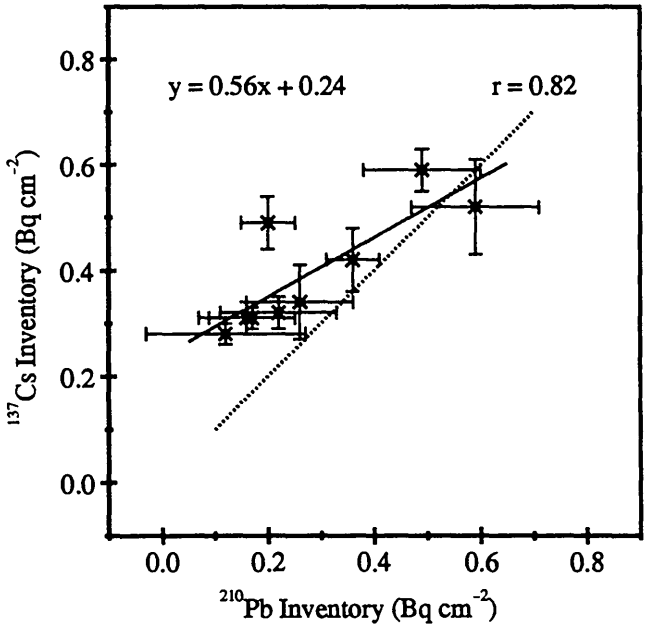

Figure 6.5: Plot of the ^{137}Cs(bomb) inventory against the ^{210}Pb(atmos) inventory for 9 sites on the western slopes of Great Dun Fell. The sites shown are G1,4,6,7,9,10,11,15,16. The solid line represents the best fit to the points and the dotted line represents the inventories increasing at the same rate

This section will discuss the levels of ^{137}Cs(Chern) found at the sampling sites and attempt to account for the pattern with altitude at each mountain and relate the values at each mountain to the picture of Chernobyl deposition to the UK as a whole. As described before, the fraction of ^{137}Cs in each counted sample due to Chernobyl is separated from the fraction due to weapons testing by using the ^{134}Cs activity of the sample. This means that the the values given for ^{137}Cs(Chern) are simply those for ^{134}Cs multiplied by the factor 1.82, ie the ratio of ^{137}Cs/^{134}Cs in rain at Eskdalemuir from April–June 1986 [23]. This section will discuss the levels of ^{137}Cs(Chern) rather than ^{134}Cs in order to make an easier comparison with the ^{137}Cs(bomb) results. However, the depth profiles presented for the Merrick will be those for ^{134}Cs.

6.4.1 Mean Values

Table 6.6 lists the mean inventory of ^{134}Cs, ^{137}Cs(Chern) and ^{137}Cs(bomb) at each of the 5 mountains. It also gives the ratio of Chernobyl derived ^{137}Cs to that due to weapons testing at each mountain. There is a great deal of variability in the levels of ^{137}Cs(Chern) between the 5 mountains, Great Dun Fell and Ben Cruachan give relatively low values of $\sim 500\,\mathrm{Bq\,m^{-2}}$ and the Merrick and Ben Lawers relatively high values in excess of $10{,}000\,\mathrm{Bq\,m^{-2}}$. The levels at Beinn Dorain are intermediate at $\sim$2,400 $\mathrm{Bq\,m^{-2}}$. In areas of the UK where there was no rainfall, during the passage of the plume, dry deposition of ^{137}Cs(Chern), to grass, amounted to only $\sim 20\,\mathrm{Bq\,cm^{-2}}$ [23][33]. Thus, it appears, that all the five locations experienced some rainfall during the passage of the plume. The levels of ^{137}Cs(Chern) at Ben Lawers are a factor of 34 greater than the levels at Great Dun Fell. It can be seen that the Chernobyl deposition is much more variable than the deposition of ^{137}Cs due to weapons testing. The difference between the mountain with the highest mean ^{137}Cs(bomb) inventory and that with the lowest is only a factor of 1.6. Table 6.6 also shows the different impact the Chernobyl accident has had at different sites. At Ben Lawers ^{137}Cs(Chern) deposition amounted to $\sim$4.5 times the amount of ^{137}Cs already present due to weapons testing. However, at

Mountain	^{134}Cs	^{137}Cs(Chern)	^{137}Cs(bomb)	Chern/bomb
Merrick	0.55 ± 0.02	1.00 ± 0.04	0.35 ± 0.04	2.9
Great Dun Fell	0.024 ± 0.002	0.044 ± 0.004	0.36 ± 0.04	0.12
Ben Cruachan	0.030 ± 0.006	0.055 ± 0.011	0.43 ± 0.08	0.13
Beinn Dorain	0.13 ± 0.02	0.24 ± 0.03	0.52 ± 0.05	0.46
Ben Lawers	0.82 ± 0.07	1.49 ± 0.13	0.33 ± 0.04	4.5

Table 6.6: Mean inventories of the radionuclides ^{134}Cs, ^{137}Cs(Chern) and ^{137}Cs(bomb) (Bq cm^{-2}) at each of the 5 mountains in this study. The uncertainties represent the standard error on the mean

Mountain	Coefficient of variation		No. of sites
	Range (%)	Mean (%)	
Great Dun Fell	6–60	29	17
Beinn Dorain	7–28	14	4
Ben Lawers	5–26	15	5

Table 6.7: Coefficient of variation, for sites at which samples were collected from 3 or more points, at Great Dun Fell, Beinn Dorain and Ben Lawers

Great Dun Fell and Ben Cruachan the Chernobyl deposition amounted to only $\sim 10\%$ of ^{137}Cs already present. These results reflect the patchy nature of the Chernobyl deposition over the UK and, with the exception of Ben Lawers, fit well with the picture of Chernobyl deposition given by Clark & Smith [33].

Clark & Smith show the areas of greatest deposition to be North Wales, Cumbria and southwest Scotland, $>20{,}000\,Bq\,m^{-2}$ in some places. However, some years after the accident it became apparent that the central Highlands also suffered high levels of ^{137}Cs deposition. It appears that Ben Lawers lies within this region of high deposition which was initially overlooked. Of the 5 mountains in this study Ben Lawers received the greatest inputs of ^{137}Cs(Chern) averaging $15{,}000\,Bq\,m^{-2}$.

Clark & Smith present a map showing the rainfall, over the UK, which intercepted the plume during 2–4 May, compiled from radar data and measurements at 4,000 rain gauge stations. The rainfall value for each location has been taken from this map and compared with the mean inventory of ^{137}Cs(Chern) to give the mean concentration of ^{137}Cs in rain during the initial passage of the plume, see Table 6.8. The range of values for the concentration of ^{137}Cs in rain-water is from 440–1,500 $Bq\,l^{-1}$, this is comparable to the 20–2,000 $Bq\,l^{-1}$, during 2–5 May 1986, reported by Smith & Clark [140]. Interestingly, the first four locations give very similar values for the rain-water concentrations, at $\sim 500\,Bq\,l^{-1}$. The exception is Ben Lawers, where the concentration of ^{137}Cs in rain-water is a factor of three larger than at the other locations. This implies that the rainfall, at Ben Lawers, during the passage of the plume was greater than is reported by Clark & Smith or, alternatively, that over Ben Lawers a section of the plume, containing a relatively high concentration of radioactive debris, was scavenged by rainfall.

In contrast to the weapons test ^{137}Cs, which was deposited over a period of several years, most of the ^{137}Cs(Chern) deposition onto the UK ground surface took place over a few days in a few rainfall events. In addition, during the passage of the plume the weather over the UK was unsettled, with heavy, localised, convective thunderstorms [140]. Because of this a greater variability in the regional

Mountain	Rain (mm)	^{137}Cs(Chern) (Bq m^{-2})	Conc. (Bq l^{-1})
Merrick	20	10,000	500
Great Dun Fell	1	440	440
Ben Cruachan	1	550	550
Beinn Dorain	5	2,400	480
Ben Lawers	10	15,000	1,500

Table 6.8: Rainfall intercepting the plume during 2–4 May 1986 [33], mean inventory of ^{137}Cs(Chern) and the calculated mean concentration of ^{137}Cs in rain during this period at each mountain location

deposition pattern would be expected. The deposition profiles at each mountain will reflect the meteorological conditions during that first week in May 1986 and these may not necessarily reflect the average conditions. The coefficient of variation was calculated for each site at which samples were collected from 3 or more points. The results for sites at Great Dun Fell, Beinn Dorain and Ben Lawers are given in Table 6.7.

6.5 Deposition Profile of ^{137}Cs(Chern) at each mountain

This section will discuss and attempt to account for the deposition profile at each of the 5 mountains.

6.5.1 Merrick

The inventory of ^{137}Cs(Chern) for each sampling site at the Merrick is shown in Figure 6.6. Pit and core values have been combined for sites M1 to M6. It is noticeable that the deposition profile is highly uniform over the mountain and

there is no evidence of an increase with altitude. Deposition of ^{137}Cs(Chern) at $\sim 10{,}000\,\mathrm{Bq\,m^{-2}}$ was relatively high at this mountain. The absence of an altitudinal increase in the ^{137}Cs(Chern) inventory indicates that scavenging of low level cloud, containing a high concentration of Chernobyl derived ^{137}Cs, was not operating during the depositional episode. There is no evidence of the deposition pattern having been influenced by the processes of seeder-feeder scavenging and occult deposition. The relatively high levels of ^{137}Cs(Chern) coupled with the uniform deposition pattern may be explained in terms of convectional rainfall events during the passage of the plume. The mean inventory of ^{137}Cs(Chern) measured at the Merrick is in line with that calculated by Clark & Smith.

The depth profiles for sites M1 to M6, see Figures 4.11 and 4.12 indicate the ^{134}Cs being present, predominantly, in the top 5 cm of the soil profile. Only at site M5 is a small amount, ie 4% of the total inventory, of ^{134}Cs observed below 10 cm depth. At site M7 no ^{134}Cs was measured below 10 cm depth and at site M8 only 4% of the inventory down to 20 cm depth was found below 10 cm, see Figure 4.13. Figure 4.13 appears to show a faster movement of ^{134}Cs down the profile at site M8 than at site M7, as was also the case for the migration of ^{137}Cs(bomb). These findings could reflect the the extremely high organic matter content at site M8, and the scarcity of illitic clay in this type of fibrous peat. It is interesting that the pit sample from site M3 gives apparently low values for the inventories of ^{210}Pb(atmos) ^{137}Cs(bomb) and ^{134}Cs, see Figure 4.1 There is no indication of down profile migration of ^{134}Cs at this site. This could indicate that either a significant fraction of rainfall flows over the location of the pit without penetrating into the soil profile or that erosion is removing small particles of soil and organic matter, with adsorbed radionuclides, from the surface of the soil.

6.5.2 Great Dun Fell

The mean deposition of ^{137}Cs(Chern) at Great Dun Fell is $\sim 440\,\mathrm{Bq\,m^{-2}}$ which is only 4% of that deposited at the Merrick. This illustrates the patchy nature of the Chernobyl deposition pattern. Cambray et al. and Livens et al. have presented

Study	^{137}Cs(Chern) ($Bq\,m^{-2}$)		No. of sites
	Lowest	Largest	
This study	150	750	23
Cambray 87	110	300	4
Livens 92	<250	570	7

Table 6.9: Comparison of 3 different sets of ^{137}Cs(Chern) measurements at Great Dun Fell

values for the ^{137}Cs(Chern) inventory in soil at Great Dun Fell [23][102]. Table 6.9 shows the range of values of the ^{137}Cs(Chern) inventory in soil at Great Dun Fell in this study and in the studies of Cambray et al. and Livens et al.. It can be seen that a similar range of values was found in each study.

Figure 6.6 shows an increase with altitude which is not smooth and monotonic but which is quite clear. The values increase from $150\,Bq\,m^{-2}$ at sites G10 and G11 to $750\,Bq\,m^{-2}$ at the summit, site G1. Thus, the results show an increase in the ^{137}Cs(Chern) inventory, between the valley and the summit, of about a factor of 5. This result would appear to indicate the operation of the seeder-feeder scavenging mechanism during the passage of the radioactive plume. However, the meteorological conditions, during the passage of the plume, were not favourable for the operation of seeder-feeder scavenging. The plume was travelling in a southeast to northwest direction and the windspeeds were generally low [102]. When airflow is from an easterly direction and the windspeed is low, enhancement of rainfall amount and wet deposition, with altitude, on the western slopes of Great Dun Fell is not observed [32][57]. In addition, rainfall during the passage of the plume was convective, rather than frontal in nature. The large enhancements in wet deposition at Great Dun Fell are associated with frontal rainfall events.

An alternative explanation is that, a convective rain shower resulted in depo-

sition of $\sim 400\,\mathrm{Bq\,m^{-2}}$ over Great Dun Fell. This is illustrated by the solid line in Figure 6.6. However, in the valley certain sites give low values of the ^{137}Cs(Chern) inventory, ie sites G7, G10 and G11. This could be due to overland flow of precipitation during deposition, or, more likely, to the patchy nature of the convectional rainfall event. Perhaps these three sites received very little rainfall during the passage of the cloud. On the contrary, at the summit, 4 sites show larger values for the ^{137}Cs(Chern) inventory, ie sites G1, G4, G19 and G20. It is possible that the values at these sites have been enhanced due to occult deposition of cloud droplets during the passage of the plume. Livens et al. argued that this was the case, to explain the high level of deposition of ^{137}Cs(Chern) they measured at the summit, in comparison to all the lower altitude sites [102].

6.5.3 Ben Cruachan

The deposition pattern of ^{137}Cs(Chern) is highly variable at Ben Cruachan, see Figure 6.6 with no clear relationship with altitude. The highest value of the ^{137}Cs(Chern) inventory is observed at site C2 ($670\,\mathrm{Bq\,m^{-2}}$) just downwind of the summit but, the lowest value at this mountain is observed at the summit itself ($90\,\mathrm{Bq\,m^{-2}}$). Deposition at this mountain was quite variable between sampling sites. It seems likely that light, patchy, convectional, showers were responsible for the patchy and, relatively, low deposition onto Ben Cruachan. It is interesting that site C9 gives a reasonable value of the ^{137}Cs(Chern) inventory when the values it gives for the ^{210}Pb(atmos) and ^{137}Cs(bomb) inventories are much too low. It could be that, the processes which have served to reduce the ^{210}Pb(atmos) and ^{137}Cs(bomb) inventories at this site, perhaps related to periodic flooding and waterlogging of the soil near the river, have not been effective in reducing the ^{137}Cs(Chern) inventory to the same extent during the four years between deposition and sampling at this mountain (20^{th} July 1990). Also, at site C5 sampling points (i) and (ii) give different values for the ^{137}Cs(Chern) inventory. This is consistent with the suggestion, put forward earlier, that erosion of topsoil is taking place at sampling point (i), thus reducing the measured inventories of

atmospherically derived radionuclides.

6.5.4 Beinn Dorain

Beinn Dorain shows a very clear increase in the ^{137}Cs(Chern) inventory with altitude, see Figure 6.7. The lowest 2 sites D10 and D11 give a mean inventory of $\sim 910\,\mathrm{Bq\,m^{-2}}$. At site D2 the inventory is $\sim 4{,}550\,\mathrm{Bq\,m^{-2}}$. The inferred deposition profile is shown by the solid line in Figure 6.7. The only site which does not follow the trend is the summit site, D1, which gives a value of the inventory which is, relatively, very low ($330\,\mathrm{Bq\,m^{-2}}$). The form of the increase with altitude is seen to be monotonic, and follows the mountain profile very closely.

The very clear increase in the ^{137}Cs(Chern) inventory, between the valley and the high altitude sites, of about a factor of 5, and the remarkable similarity between the form of the deposition profile and the shape of the mountain itself, makes Beinn Dorain the most interesting of the 5 mountains in terms of Chernobyl deposition. In the absence of detailed knowledge of the meteorology during deposition, it is difficult to identify, with confidence, the processes producing the observed pattern. Seeder-feeder scavenging could be invoked to explain the factor increase in deposition and the form of the deposition profile but, as was explained for Great Dun Fell, conditions during the passage of the main body of the plume (2–4 May) were not favourable for the operation of this mechanism.

A second possible mechanism is that of occult deposition. Cloud droplets, of radius 2.5–15.5 μm, typically show deposition velocities of magnitude $\sim 60\,\mathrm{mm\,s^{-1}}$ [63]. The concentration of ^{137}Cs in air during 2–3 May 1986 at Chilton was measured to be $0.5\,\mathrm{Bq\,m^{-3}}$ [23]. A cloud, containing this concentration of ^{137}Cs, could deposit $\sim 2{,}000\,\mathrm{Bq\,m^{-2}}$ in a 20 hour period, which is about half the period of duration of the plume at Great Dun Fell [102]. Thus, it appears that occult deposition of cloud droplets could result in a deposit of ^{137}Cs(Chern) of the same magnitude as that measured, in soil, at the mountain. However, it is difficult to account for the high deposit of almost $5{,}000\,\mathrm{Bq\,m^{-2}}$, at site D2, by invoking occult deposition, unless the concentration of ^{137}Cs in air at 1,000 m altitude was greater than that

measured at ground level at Chilton. Another problem with this explanation, is that, the increase of ^{137}Cs(Chern) with altitude is so smooth. If occult deposition were responsible for the observed pattern, an abrupt change in the slope of the deposition profile would be expected. This would correspond to the altitude of the mean cloud base, with the ground above cloud base receiving an input of cloud droplets, containing ^{137}Cs, and that below cloud base receiving no such input.

An alternative explanation is that Beinn Dorain experienced a strong, local, convectional rainstorm event during the passage of the plume. Heating of the ground surface may have lead to strong vertical updrafts of warm, moist air centred over the summit of the mountain. This may have generated rain, with the highest rates falling onto the summit and progressively lower rates falling with distance from the summit. The low value at site D1 could be due to overland flow during the deposition, ie rainfall running over the surface with little percolation into the soil profile. The observed deposition profile of ^{137}Cs(Chern) at Beinn Dorain is of great interest as it shows a very clear altitude dependence. Unfortunately, due to the lack of detailed knowledge of the meteorology during deposition, the interpretation is very speculative.

6.5.5 Ben Lawers

Of the 5 mountains, the greatest deposition of ^{137}Cs(Chern) was found at Ben Lawers, averaging $\sim 15{,}000\,\mathrm{Bq\,m^{-2}}$. The deposition profile is interesting at this mountain as the values are very uniform from site L15 to site L6. At higher altitudes than site L6 the deposition pattern becomes highly variable, see Figure 6.7. The high variability could be due to complex high altitude meteorology at Ben Lawers. Alternatively, post-depositional processes could be responsible. If post-depositional processes are responsible then a possible interpretation could be that the values at sites L1,L2 and L5 are low due to overland flow and that site L4 is situated in an area of ground which received overland flow during the deposition episode. Below 1,000 m altitude the deposition pattern of ^{137}Cs(Chern) is very even, indicating that the rainfall rate was very consistent, over this large altitude

range, during the passage of the plume. It is possible that the main body of the plume, maybe at $\sim$1,000 m altitude, passed around, rather than over, the upper $\sim$200 m of Ben Lawers. Rainfall, from above, falling onto the highest reaches of the mountain would not have fallen through the main body of the plume but a more patchy, thinner part of it.

6.6 The ^{7}Be Inventory at the Merrick

The radionuclide ^{7}Be was measured in samples taken during the first expedition to the Merrick (sites M1–M6). At each of these six sites three sets of cores were taken and a single pit was dug. In addition, samples of vegetation were taken from sites M3 to M6, but there was insufficient vegetation present at the highest two sites. At sites M3 to M6 vegetation was clipped from three one metre squares and the pit was then taken from within one of these squares. No ^{7}Be was measured below 1 cm depth in the cores or below 5 cm depth in the pits. At these six sites, ^{7}Be is retained very close to the ground surface, within $\sim$1–2 cm depth, and does not penetrate very far down the soil profile. As ^{7}Be has a relatively short half-life of 53.3 days it is expected to be present within a few cm of the surface. Olsen et al. took soil cores at Wallops Island, Virginia and found that ^{7}Be is confined to the surface when the marsh soils are saturated but may penetrate to depths as great as 10 cm when the marsh soil is unsaturated [119]. Rapid infiltration of rainwater could account for the relatively deep penetration when the soil is unsaturated.

Whereas the amounts of ^{210}Pb(atmos) and ^{137}Cs adsorbed to vegetation were found to be insignificant, compared to the inventory present in the soil profile, this is not the case for ^{7}Be. Because of its relatively short half-life a significant fraction of the ^{7}Be inventory is found to be associated with the vegetation cover. Table 6.10 gives the fraction of the total ^{7}Be found to be associated with the vegetation cover. The vegetation cover at sites M3 to M5 was mostly rough grass while at site M6 bracken *(Pteridium aquilinum)* was present.

The measured inventories from the pits and cores are shown in Figures 4.2

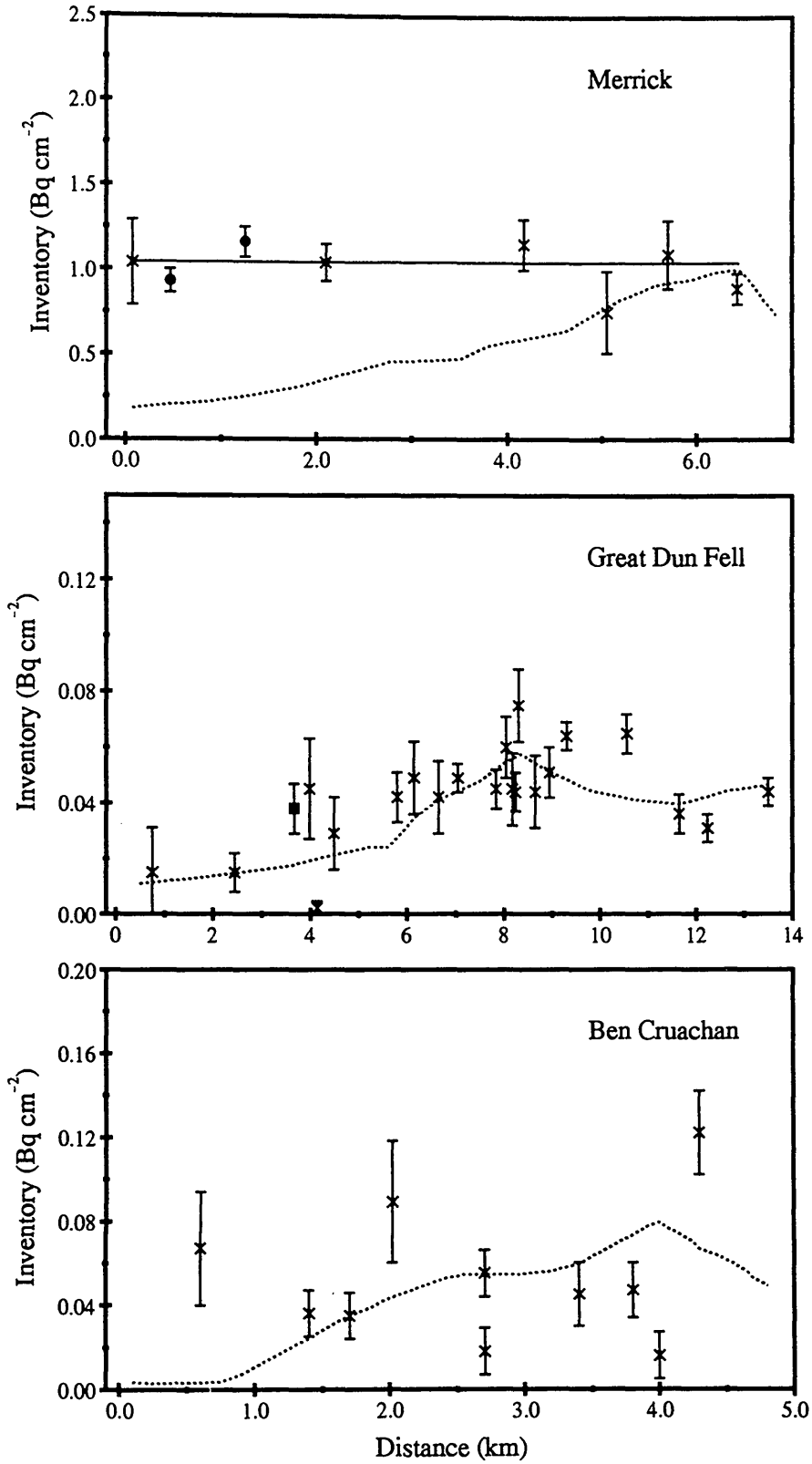

Figure 6.6: Inventory of ^{137}Cs(Chern), Bq cm^{-2}, at the Merrick, Great Dun Fell and Ben Cruachan. The solid line represents the inferred deposition profile

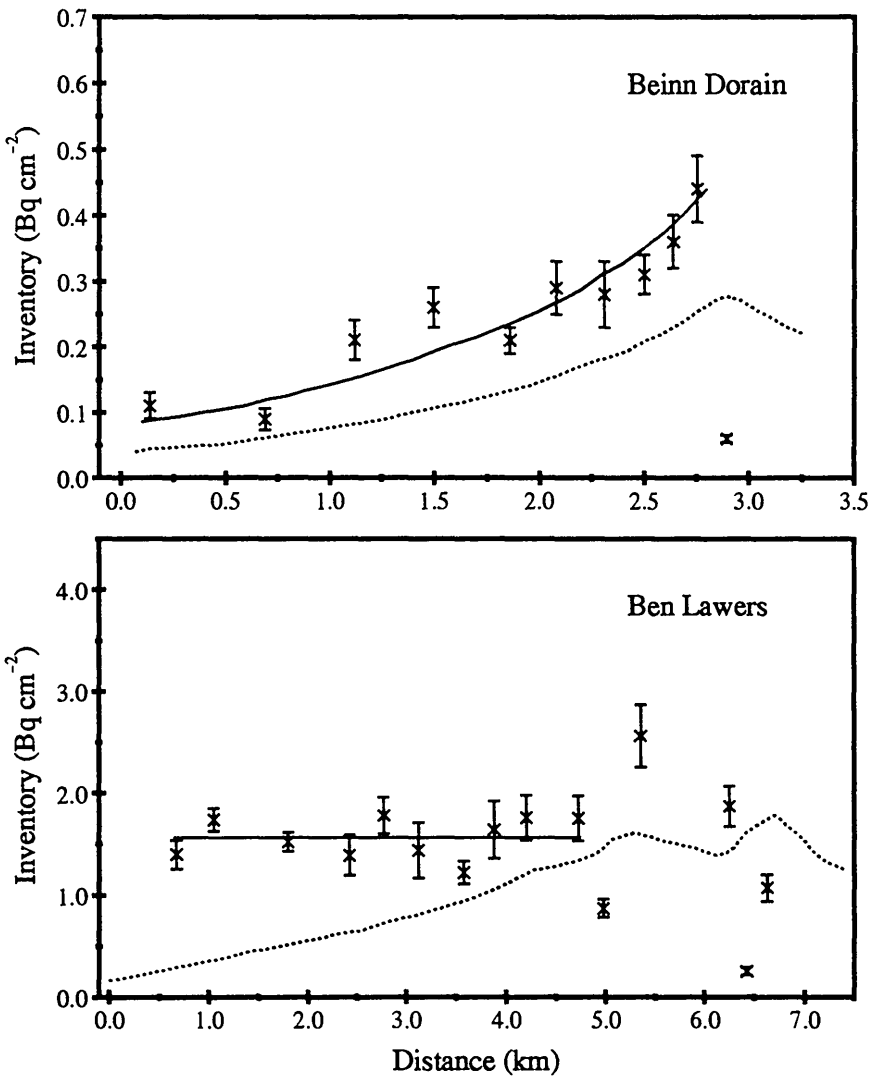

Figure 6.7: Inventory of ^{137}Cs(Chern), Bq cm^{-2}, at Beinn Dorain and Ben Lawers. The solid line represents the inferred deposition profile

Site	M1	M2	M3	M4	M5	M6
%	-	-	44	20	41	100

Table 6.10: Percentage of the total ^{7}Be Inventory associated with the vegetation at sites M3 to M6

and 4.4 respectively. A clear increase with altitude is seen in both cases of a factor ~ 2. The error margins for the core and pit values overlap except at sites M3 and M4. These two independent sets of values have been combined in Figure 6.8. The error bars represent the standard error from the combination of the 2 separate sets of values or the combination of the individual errors, whichever is the greatest. With the ^{7}Be data presented here an increase in deposition with altitude is observed which is greater than the increase in the mean annual rainfall with altitude. This observation could be attributed to the processes of seeder-feeder scavenging and occult deposition which act to scavenge boundary layer aerosols very efficiently over mountains. However, it is not valid to make a comparison with the mean annual rainfall pattern as the measured inventories of ^{7}Be, probably, do not represent the mean annual flux from the atmosphere.

The half-life of ^{210}Pb (22.3 years) is long in comparison to the timescale of seasonal changes in the ^{210}Pb(atmos) flux to the ground, which are probably in the order of $\sim$2–3 months. Because of this, the mean annual flux to the ground surface, averaged over several decades, may be obtained from the steady state inventory in the soil. In contrast, the short half-life of ^{7}Be (53.3 days) is less than the timescale of seasonal changes in the flux to the ground surface, five half-lives of ^{7}Be corresponds to a period of ~ 260 days. Thus, the ^{7}Be inventory on the ground will reflect the flux to the ground during the preceding few months. Many authors, eg. [43][119], have presented results which show seasonality in the ^{7}Be flux to the ground. On the eastern coast of the USA the flux is usually a maximum in the spring and a minimum in the autumn. The results from sites M1 to M6 give

Reference	Location	Measurement	Dates	^{7}Be Inventory ($Bq\,cm^{-2}$)
Peirson [125]	Chilton, UK	rain	1960	0.020
	Milford Haven, UK	rain	1960	0.032
Olsen et al. [119]	Oak Ridge, USA	soil & veg	1982–84	0.037–0.074
Casey et al. [25]	Norfolk, USA	soil & veg	1984–85	0.064
Turekian	New Haven, USA	rain	1977–78	0.080
et al. [146]	Bermuda	rain	1977–78	0.060
This study	Galloway, UK	soil & veg	Oct. 1988	0.039

Table 6.11: Values of the ^{7}Be inventory for sites in the UK and the USA, either measured directly in soil and vegetation or inferred from measurements in rainfall

a mean value of $0.039 \pm 0.006\,Bq\,cm^{-2}$ for the ^{7}Be inventory. This is within the range of values found in the literature from direct measurements of the inventory and from measurements of ^{7}Be in rainfall see Table 6.11.

The atmospheric flux of ^{7}Be is expected to vary less within the same band of latitude than the ^{210}Pb(atmos) flux. Whereas ^{210}Pb(atmos) is derived from continental land masses the production rate of ^{7}Be is the same over land and sea. There is evidence that the flux of ^{210}Pb(atmos) is greater to the east coast of the USA than to the UK as a result of westerly flow of air over the Atlantic ocean [30][145].

If there is a seasonal variation in the ^{7}Be flux to the UK then it may be expected that the steady state inventory on the ground will vary significantly through the course of the year. Todd et al. measured ^{7}Be in rain over a 29 month period at Norfolk, Virginia [143]. From their measurements, they calculated that the steady state inventory of ^{7}Be varied by a factor of ~ 2 through the year with a minimum during October.

Sampling at the Merrick took place during October 1988 so it is possible that

the ^{7}Be inventory is different at other times of the year. A greater inventory may be present during the spring. However, the measurements reported by Peirson showed that, while the concentration of ^{7}Be in surface air and rainfall was greatest in the spring, high rainfall during the autumn and winter meant that the flux of ^{7}Be in rainfall was greatest during these months [125].

Measurement of ^{7}Be offers some interesting opportunities for further work. ^{7}Be is present on vegetation and within a few cm of the ground surface, so large quantities of soil need not be taken to measure the inventory. If samples were taken at different altitudes and at regular intervals throughout the year (eg. every month or two months) then information may be obtained on seasonal changes in the pattern of deposition with altitude. The different processes, responsible for deposition from the atmosphere, may vary in importance throughout the year, for example convectional rainfall may be important during the summer months.

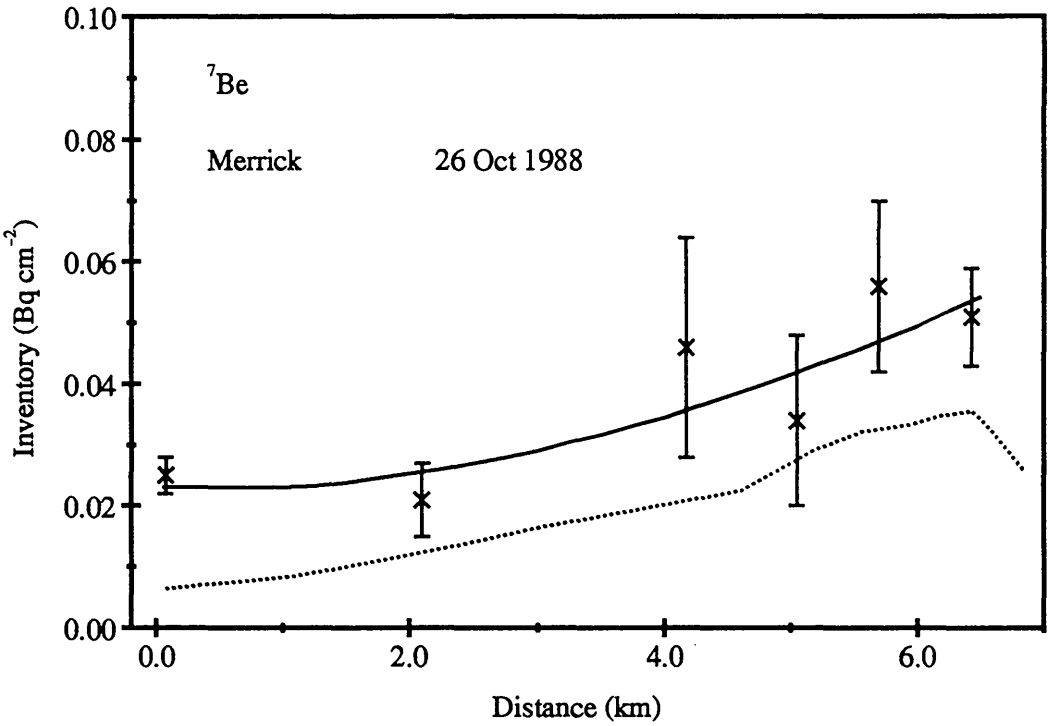

Figure 6.8: The ^{7}Be Inventory (Bq cm^{-2}) measured at the Merrick on 26^{th} Oct 1988. Sites M1 to M6, pit and core results combined. Solid line represents the inferred deposition profile

Chapter 7

Conclusions

The main objective of this study has been to measure atmospheric derived ^{210}Pb in soil and establish the influence of altitude and rainfall on deposition at a range of mountain locations in northern Britain.

Lead-210 in the atmosphere is attached to aerosols which are deposited to the ground by precipitation scavenging, turbulent deposition of cloud droplets and dry deposition. The results of the ^{210}Pb(atmos) investigation have been used to infer deposition patterns of acidic ions, SO_4^{2-} NO_3^- etc, in areas of complex topography, as these ions are associated with the same sub-micron aerosols as ^{210}Pb, and also as a vaildation of the modifications made to the map of wet deposition in order to account for the effect of seeder-feeder scavenging [47]. Other radionuclides, ^{137}Cs, ^{134}Cs and ^{7}Be have also been measured, in the same soil samples as the ^{210}Pb and results presented and discussed. The following conclusions summarize, the advantages and disadvantages of the soil sampling technique, increases in ^{210}Pb deposition with altitude (and, by implication, deposition of acidic ions), changes in the deposition pattern along the Highland transect, implications for the modified wet deposition map of the UK [47], the ^{137}Cs(bomb), ^{137}Cs(Chern) and ^{7}Be results and an assessment of the radiological hazard associated with the levels of radiocaesium at Ben Lawers.

Advantages. The soil coring technique has proved to be cheap, simple and effective. Taking samples in the field is quite straightforward, requiring very little training and several sites may be sampled during the course of one day. At the majority of sites the bulk of the ^{210}Pb(atmos) inventory was found to reside in the top 10 cm of the soil profile. Sampling involves little cost as no expensive equipment is required. The technique provides a relatively detailed indication of the deposition field and its spatial variability, as a large number of sites may be sampled (66 in this study). In comparison, experiments which involve measuring the deposition of ions in precipitation, eg SO_4^{2-}, NO_3^-, H^+, NH_4^+ etc, require relatively frequent attention with work usually being carried out on an event basis. Thus the soil coring technique has a greater versatility, in terms of the number of sites which may be sampled and the fact that areas inaccessible to chemical monitoring may be sampled. Chemical measurements give a very detailed picture of depositional processes during indiviudual meteorological events, ie few hours or days duration, with measurements being made at a small number of monitoring sites. The measurement of ^{210}Pb in soil, in contrast, allows a larger scale picture of the wet deposition field to be gained. Also, the ^{210}Pb technique integrates deposition over many decades thus allowing the identification of processes which influence the total deposition pattern. If the ^{210}Pb(atmos) inventory in soil represents deposition during the previous ~ 50 years then, with with say 150 precipitation events a year, the inventory will represent the integrated deposit due to several thousand events The ^{210}Pb technique thus complements the detailed chemical investigations of atmospheric deposition, eg at Great Dun Fell, by extending the study of deposition patterns, in mountain areas, both temporally and spatially.

Disadvantages. The technique does however have some disadvantages. Although great care has been taken, one area of uncertainty which cannot be remedied is the fact that the character of the collecting medium, ie the soil, varies. Soil type changes between the valleys and the hilltops in this study. Another draw-

back is the time required to analyse the samples by γ-ray spectroscopy. Many individual soil samples are produced and each individual sample needs on average 1 day for counting. Ideally several detectors are rquired in order to speed up the analysis.

The pilot study at the Merrick indicated the importance of consistent field sampling procedure and detector performance stability during counting. The values from the original six sampling sites (M1–M6) show an increase in the ^{210}Pb(atmos) inventory with altitude. However, sampling at the two later sites (M7 and M8) suggests that the original values are systematically low. The results from this location must therefore be treated with some caution. The sampling technique was improved during the course of the study, with greater subdivision down the cores and the practice of keeping material from individual sampling points separate. For each location the coefficient of variation was calculated for each site at which three or more points were sampled. The mean coefficient of variation for each location was 34% at the Merrick, 29% at Great Dun Fell, 14% at Beinn Dorain and 18% at Ben Lawers. This decline has been due largely to improvements in the sampling procedure during the course of the study.

^{210}Pb inventories in soils

The mean inventory of 65 sites (ie excluding site C9) was $0.44 \pm 0.03\,\mathrm{Bq\,cm^{-2}}$, the uncertainty represents the standard error of the mean. The mean of the average annual rainfall for these 65 sites, as given by the average rainfall map, is $2{,}060 \pm 70\,\mathrm{mm\,yr^{-1}}$. Thus the standard deviation associated with the mean ^{210}Pb(atmos) inventory is greater than that associated with the mean annual rainfall. These values correspond to a mean concentration in rainfall of $\sim 66\,\mathrm{mBq\,l^{-1}}$. This compares very closely with the concentration in rainfall measured by other workers, eg Eakins & Morrison $74\,\mathrm{mBq\,l^{-1}}$. This is especially so if we assume that the soil inventory of ^{210}Pb(atmos) underestimates the actual input by 10% [149].

Six sites at the base of Great Dun Fell give a mean inventory of $0.19\,\mathrm{Bq\,cm^{-2}}$

with 1,040 mm yr^{-1}. This was used as the baseline value for this location. In order to obtain a value of the ^{210}Pb(atmos) inventory at the western coast of Scotland, ie upwind of the Highland mountain mass, the values from sites C7 and C8 were extrapolated to 1,000 mm yr^{-1} rainfall and the extrapolated inventory was 0.17 Bq cm^{-2}. This was used as the baseline value for the Highland Transect. These two baseline values are close to that of 0.22 Bq cm^{-2}, with 1,100 mm yr^{-1}, measured at Plymouth by Clifton [35].

Increases in deposition with altitude

The effects of altitude on the deposition of ^{210}Pb will now be summarised. Of the 5 locations, the ^{210}Pb(atmos) inventory shows a clear increase clearly with altitude at 4 (Merrick, Great Dun Fell, Ben Cruachan and Beinn Dorain). The increases are greater than the increase in rainfall indicated on the 30 year mean annual rainfall map. At the remaining location, Ben Lawers, the results may indicate a slight increase in the inventory with altitude, but, the rate of increase is not significantly greater than that of the mean annual rainfall.

The increases in the inventories at Great Dun Fell, Ben Cruachan and Beinn Dorain between the valley and the summit are; 3.3, 2.8 and 2.4 respectively. These compare with rainfall increases of 2, 1.7 and 1.2 respectively. The large excess deposition of ^{210}Pb, above that of rainfall, is entirely consistent with the effects of seeder-feeder scavenging and occult deposition which both operate to scavenge the low level cloud which frequently shrouds the upper reaches of these mountains. On average, the upper slopes of these mountains receive rainfall which contains a concentration of ^{210}Pb which is about $\times$ 1.5 that found in the seeder rainfall. This would correspond to cloud water concentrations of ^{210}Pb at these 3 mountains being, on average $\times$ 2 that of the seeder rainfall. At the summit of Great Dun Fell the results indicate that cloud water contains a concentration of ^{210}Pb which is $\times$ 2.6 that in the seeder rain. These values correspond very closely with those given for deposition of the major ions at Great Dun Fell [57].

The deposition profiles of ^{210}Pb at Great Dun Fell and Beinn Dorain are best

explained by including a significant additional contribution by occult deposition at their summits. It is estimated that occult deposition is responsible for 23% of total deposition at the summit of Great Dun Fell and at least 30% of deposition at the summit of Beinn Dorain. These values are higher than those of other workers, eg [45][59][77]. This could be due to the altitude of the measurements in this study. Occult deposition rates increase with altitude, due to the higher windspeeds and increased cloud immersion frequency. The occult deposition inputs calculated by these workers have been for altitudes lower than the altitude of soil sampling in this study.

The deposition profile at Great Dun Fell is consistent with high rates of occult deposition at the summit. This is in contrast to the model of Hill et al. which predicts the highest rates of occult deposition of sulphate to be downwind of the hill summit [80]. This is due to the increase in windspeed downwind of the hill summit when the airflow is considered to be supercritical. However, when the airflow pattern and stability leads to subcritical flow over parts of the downslope the highest mean windspeeds and thus sulphate deposition rates are found at the summit.

The calculated enhancements are consistent with those measured in chemical experiments, Fowler et al. [57] found cloud water to contain concentrations of the major ions typically 2–8 times higher than in the seeder rainfall, and with the predictions of a model of the deposition of acidic ions [32]. The results of this study have been used to deduce the concentrations of ^{210}Pb in low level cloud droplets in comparison to those in the seeder rain. Around the summits of Ben Cruachan, Beinn Dorain and Great Dun Fell, cloud water contains concentrations of ^{210}Pb typically $\times 2$ those in the seeder rainfall. Larger values found in the chemical experiments could be a result of in-cloud oxidation (for sulphate), episodes with very polluted boundary layer air and also due to the fact that chemical experiments have been conducted under conditions where the seeder-feeder scavenging mechanism is operating. In contrast, the ^{210}Pb(atmos) inventory in soil represents the atmospheric flux integrated over many decades, and thus several thousand

precipitation events. Thus, the inventory in soil records the deposition under all meteorological conditions, ie including dry deposition and deposition in rainfall in the absence of seeder-feeder scavenging. The measured deposition profile indicates that seeder-feeder scavenging and occult deposition are significant contributors to the long-term deposition pattern.

Attributing the enhancements with altitude in ^{210}Pb deposition to seeder-feeder scavenging (and occult deposition) implies that boundary layer air is enriched in ^{210}Pb relative to levels in the lower troposphere where the bulk of the seeder rain is generated. As the source of ^{210}Pb is radon emanating from the ground surface, this seems a reasonable assumption to make. However, measurements of the vertical profile of ^{210}Pb concentration in the troposphere show the concentration of ^{210}Pb to increase with altitude over the UK [21][127]. Although this may be true over the full height of the troposphere, it is likely that, for the boundary layer, and close to the surface, the concentration of ^{210}Pb(atmos) is greater than in the overlying lower troposphere. Many airflows are from the east and are likely tobe rich in ^{210}Pb. This is likely to be the consequence of European 'contaminated' air being advected over the UK as is clearly the case for SO_4^{2-}, NO_3^- etc [57].

Wet deposition in complex terrain. The Highland Transect

The Highland Transect involved sampling at 3 mountains, the first Ben Cruachan, close to the western coast, Beinn Dorain and Ben Lawers which is situated ~ 80 km inland. At Ben Cruachan and Beinn Dorain the ^{210}Pb(atmos) inventory increases with altitude at a greater rate than does the rainfall, but at Ben Lawers the increase is no greater than that for rainfall.

The mean ^{210}Pb inventory at each mountain, and the peak inventory, decreases inland from Ben Cruachan to Beinn Dorain to Ben Lawers at a greater rate than rainfall amount decreases. This can be explained as a 'cleansing' of boundary layer air as it is transported inland from the coast, as observed by Dore et al. [46]. This 'cleansing' involves a progressive removal of particulate from the lower

atmosphere, by rainfall scavenging, as the precipitating airmass passes over successive hills. In addition, Ben Lawers is sheltered to the west by an extensive area of mountains, and its topographic situation is complex. Also weather originating from the east becomes more significant towards the centre of Scotland. In summary, the observations suggest a progressive weakening of the importance of low level cloud in the overall deposition pattern with distance inland from the coast. This is due, in the main, to the 'drying' and 'cleansing' of low level air during passage over successive mountains. However, the topographic situation at Ben Lawers, and also possibly at Beinn Dorain, is complex and the deposition pattern with altitude here will also reflect this.

The UK wet deposition map

Dore et al. modified the wet deposition map of the UK, to take account of the seeder-feeder effect, by assuming that all rainfall in excess of that falling at the coast is produced by the scavenging of low level cloud droplets [47]. They assumed that these low level cloud droplets contain concentrations of the major ions which are, on average, double those in the seeder rain. Using this technique, estimates of wet deposition in some upland areas increased by up to 76%.

The ^{210}Pb(atmos) results from Great Dun Fell, Ben Cruachan and Beinn Dorain imply a concentration of ^{210}Pb in low level cloud, on average, twice that in seeder rain. Assuming that SO_4^{2-} and ^{210}Pb have similar distributions, behave similarly in the atmosphere and are subject to the same deposition processes then the results of this study provide strong support for the approach of Dore et al.. The results from the Highland Transect seem to show a decline in the enhancement of ^{210}Pb(atmos) in soil with distance inland, due to 'cleansing' of the airmasses, suggesting that possibly the modified map overestimates wet deposition inland. However, there are important source differences between ^{210}Pb and, for instance, SO_4^{2-}. The modified map may indeed be slightly overestimating wet deposition for inland remote areas such as the north of Scotland, but there are many large scale sources for the acidic ions in inland areas of the UK, eg large cities such

as Manchester located between the coast and high ground. In this case it is expected that airmasses travelling across the country will encounter these additional sources of acidic ions, thus, in some degree, compensating for the 'cleansing' by rainfall. It is possible, therefore, that the map of Dore et al. underestimates wet deposition to uplands which are close to major pollution sources, eg the southern Pennines.

Further work

This study has focused on sampling at individual mountains. It would be instructive to set these individual mountains in a wider context, for instance extending the Highland Transect to the west, perhaps sampling on the Western Isles, and to the east coast, perhaps around Aberdeen. This would produce baseline values for the western and eastern extremeties of the transect. It would also be instuctive to carry out sampling in other countries.

The technique has been shown to be cheap, simple and effective, although time consuming in terms of the γ-ray analysis. It is possible for samples to be taken over extensive areas to help define the wet deposition field over larger areas to check and extend the present maps of deposition. This could possibly involve the collaboration of groups at other research centres.

On a smaller scale the technique could be used to examine the effects of topographic features on deposition. The technique could be used to examine the effect of valleys dissecting a highland mass on the deposition pattern. Valleys which lie parallel to the dominant airflow are liable to 'funnel' air whereas narrow valleys lying perpendicular to the airflow may be sheltered [123]. It might also be possible to examine the effect of slope on the deposition pattern. There is evidence at Ben Lawers that deposition increases where the gradient of the slope is greatest. Another area to investigate may be the effects of forests which scavenge low level air very efficiently.

This study and these suggested studies would be complemented by measurements of ^{210}Pb in rain and also in air. Knowledge of the vertical distribution

of ^{210}Pb in the lower part of the troposphere would be very useful in the interpretation of the values of ^{210}Pb(atmos) in soil. It is necessary to determine the concentration of ^{210}Pb in boundary layer air in comparison to that in the troposphere above, where the seeder rainfall originates. For instance, if the concentration of SO_4^{2-} in air decreases, with height, at a greater rate than ^{210}Pb then we would expect the enhancement in deposition with altitude, due to seeder-feeder scavenging, to be greater for SO_4^{2-} than for ^{210}Pb. It is also important to measure the variability in air concentrations of ^{210}Pb with wind direction.

Large uncertainties exist in the precipitation field (in the order of 30% on mountain tops) so that, if rainfall at the mountains of interest were more accurately measured, or estimated, this would greatly help in the comparison of ^{210}Pb(atmos) increases with altitude with the increases in rainfall with altitude.

From the same soil and vegetation samples collected for ^{210}Pb analysis, ^{137}Cs and ^{7}Be were also measured. The ^{137}Cs in each sample was separated into the fraction from the weapons testing, ^{137}Cs(bomb), and the fraction due to Chernobyl, ^{137}Cs(Chern).

^{137}Cs(bomb) inventories

The mean ^{137}Cs(bomb) inventory in soil, ranged from 3,300 Bq m^{-2} at Ben Lawers to 5,200 Bq cm^{-2} at Beinn Dorain. The average depth of sampling at the 5 mountains was 15 cm and it is evident that a significant fraction of the atmospheric input has not been retained within this depth range. Based on the relationship of Cawse & Horill between ^{137}Cs(bomb) in soil and rainfall it appears that $< 60\%$ of the atmospheric input has been measured [27]. For individual sites, the measured values of ^{137}Cs(bomb) are as low as 12% of that expected (site C9).

Some sites posess ^{137}Cs(bomb) inventories which are much higher than those at neighbouring sites, and are also high compared to the amount expected from the mean annual rainfall. It is thought that these sites are receiving inputs of ^{137}Cs(bomb) from surrounding areas, possibly on eroded fine particles or due to focusing of overland flow during rainfall events. It is apparent that ^{137}Cs is

relatively mobile in the upland organic soils which characterise many of the soils in the study area. In order to use the soil inventory of ^{137}Cs(bomb) to investigate the atmospheric deposition pattern in such areas it is essential to sample deeper than 15 cm, preferably to bedrock, and also to avoid fibrous peat which tends to act as a slow release source for ^{137}Cs [81][85].

The core samples from sites M1 and M2 at the Merrick may indicate high rates of leaching/translocation, of ^{137}Cs, down the soil profile at the summit of this mountain. Forty-two percent of the expected ^{137}Cs(bomb) input was measured in the cores from sites M1 and M2 compared to 82% at sites M3 and M4. This could possibly be related to high rainfall at the summit.

Because of lateral movement and migration down the profile, the ^{137}Cs(bomb) measurements are largely unsuitable for the investigation of atmospheric deposition processes. However, ten sites at Great Dun Fell appear to have retained the atmospheric input of ^{137}Cs(bomb) within the sampling depth. For these sites the ^{210}Pb(atmos) and ^{137}Cs(bomb) inventories have been compared. The ^{210}Pb(atmos) inventory increases with altitude at a faster rate than rainfall, whereas the ^{137}Cs(bomb) inventory appears to increase at the same rate as the rainfall. This implies that the vertical distribution of ^{137}Cs(bomb) in the atmosphere during deposition was uniform with altitude. In contrast, the relationship of the ^{210}Pb(atmos) inventory with altitude suggests that the concentration of ^{210}Pb in the atmosphere decreases with altitude from the boundary layer to the middle troposphere. The results suggest that ^{210}Pb is enriched in boundary layer air whereas ^{137}Cs(bomb), during the time of deposition, was not.

The decrease in the ratio ^{137}Cs(bomb) / ^{210}Pb(atmos) with altitude at Great Dun Fell has been observed in the United States and is considered to indicate the preferential scavenging of boundary layer air by mountainous terrain [73][110].

^{137}Cs(Chern) inventories

The ^{137}Cs and ^{134}Cs from the Chernobyl reactor failure was largely deposited during one or two days. Thus, the pattern of deposition reflects the meteorological

conditions during these few days, unlike the ^{210}Pb(atmos) and ^{137}Cs(bomb) (where retained) inventories which represent deposition integrated over several years at least. The deposition was highly variable between locations, in contrast to the situation for ^{137}Cs(bomb) deposition. The ^{137}Cs(Chern) levels vary from only 12% of ^{137}Cs(bomb) already present, at Great Dun Fell, to $\times 4.5$ that already present at Ben Lawers. The mean ^{137}Cs(Chern) inventory at Ben Lawers is a factor of 34 greater than that at Great Dun Fell. This picture is consistent with the very patchy nature of deposition onto the UK, through convective rainstorms, and excepting Ben Lawers, the inventory at each mountain is in line with that indicated by Clark & Smith [33]. At Ben Lawers the unexpected high levels suggest either that rainfall was greater than given or that the concentration of radioactivity in the portion of the plume which passed over this mountain was greater than the concentration in the rest of the plume.

The highest values of the ^{137}Cs(Chern) inventory, $\geq 10{,}000\,\mathrm{Bq\,cm^{-2}}$ were found at Ben Lawers and the Merrick and the deposition pattern was very uniform at these two mountains. It is thought that light showers were responsible for the relatively low inventories at Great Dun Fell and Ben Cruachan. At Beinn Dorain a large and monotonic increase in the ^{137}Cs(Chern) inventory with altitude was observed, of about a factor of 5 between the valley and the summit. Conditions were not favourable for the operation of the seeder-feeder scavenging mechanism and the values appear to be too high to be due to occult deposition alone. Strong convective updraughts, centred on Beinn Dorain itself and leading to rainfall, may have been responsible for the remarkable deposition pattern at this mountain.

Radiological hazard of radiocaesium in the ground

It is instructive to make a rudimentary assessment of the radiological hazard due to the relatively high levels of man-made radiocaesium at the locations featured in this study. At Ben Lawers, the inventories of ^{137}Cs and ^{134}Cs are $18{,}000\,\mathrm{Bq\,m^{-2}}$ and $8{,}200\,\mathrm{Bq\,m^{-2}}$ respectively. Using the equation of Chamberlain [28], which relates the dose rate at 1 m above an infinite plane surface covered with a con-

centrations of specific radionuclides, a person spending a third of the year on Ben Lawers would be at risk of receiving a dose of $0.29\,\mathrm{mSv\,yr^{-1}}$ from these two radioisotopes. (The assumption of an infinite plane surface means that this figure will overestimate the actual dose rate.) This value is equivalent to 16% of the average background dose rate per person, $1.87\,\mathrm{mSv\,yr^{-1}}$, due to natural sources. Thus, a person spending a third of the year out of doors would receive a dose, from radiocaesium in the ground, which is a small proportion of the natural background dose, but which is not an insignificant amount.

^{7}Be measurements

Beryllium-7 was measured in samples from Merrick sites M1-M6, sampled on 26^{th} Oct 1988. The measured inventory shows an increase of about a factor of 2 between the valley and the summit. The relatively short half-life (53.3 days) and the fact that the rainfall distribution, with altitude, responsible for depositing the ^{7}Be is not known, means that the results are of limited value for the purposes of this study. The mean of the measured inventories is $0.039\,\mathrm{Bq\,cm^{-2}}$ which, if the inventory were in steady state equilibrium with the atmospheric flux, would correspond to a depositional flux of $0.5\,\mathrm{mBq\,cm^{-2}\,d^{-1}}$. This value is within the range of values measured by other workers [119][125]. The short half-life means that ^{7}Be has potential for use as a tool to investigate seasonal variations in the deposition pattern, and also the influence of large precipitation events.

Summary

The main aim of this thesis has been to measure ^{210}Pb and use it as a tracer of the wet deposition field in areas of complex topography. The results of the sampling at Great Dun Fell compare very favourably with the measurements of SO_4^{2-} and NO_3^- deposition [57] and a model of wet deposition over this mountain [32]. Sampling along the Highland Transect has produced a valuable set of data on the changing deposition field with distance inland from the western coast. Qualitatively the results are as might be expected from the work of Dore et al.

[46] on measurements of the deposition field over complex terrain and support the assumptions made by Dore et al. [47] in revising the map of wet deposition for the UK, namely in the assumption that cloud water is twice as concentrated as seeder rainfall.

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